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REVIEW



## Advances and prospects in the food applications of pectin hydrogels

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### ABSTRACT

Pectin hydrogel is a soft hydrocolloid with multifaceted utilities in the food sector. Substantial knowledge acquired on the gelation mechanisms and structure-function relationship of pectin has led to interesting functions of pectin hydrogel. Food applications of pectin hydrogels can be categorized under four headings: food ingredients/additives, food packaging, bioactive delivery and health management. The cross-linked and tangly three-dimensional structure of pectin gel renders it an ideal choice of wall material for the encapsulation of biomolecules and living cells; as a fat replacer and texturizer. Likewise, pectin hydrogel is an effective satiety inducer due to its ability to swell under the simulated gastric and intestinal conditions without losing its gel structure. Coating or composites of pectin hydrogel with proteins and other polysaccharides augment its functionality as an encapsulant, satiety-inducer and food packaging material. Low-methoxyl pectin gel is an appropriate food ink for 3D printing applications due to its viscoelastic properties, adaptable microstructure and texture properties. This review aims at explaining all the applications of pectin hydrogels, as mentioned above. A comprehensive discussion is presented on the approaches by which pectin hydrogel can be transformed as a resourceful material by controlling its dimensions, state, and rheology. The final sections of this article emphasize the recent research trends in this discipline, such as the development of smart hydrogels, injectable gels, aerogels, xerogels and oleogels from pectin.

### KEYWORDS

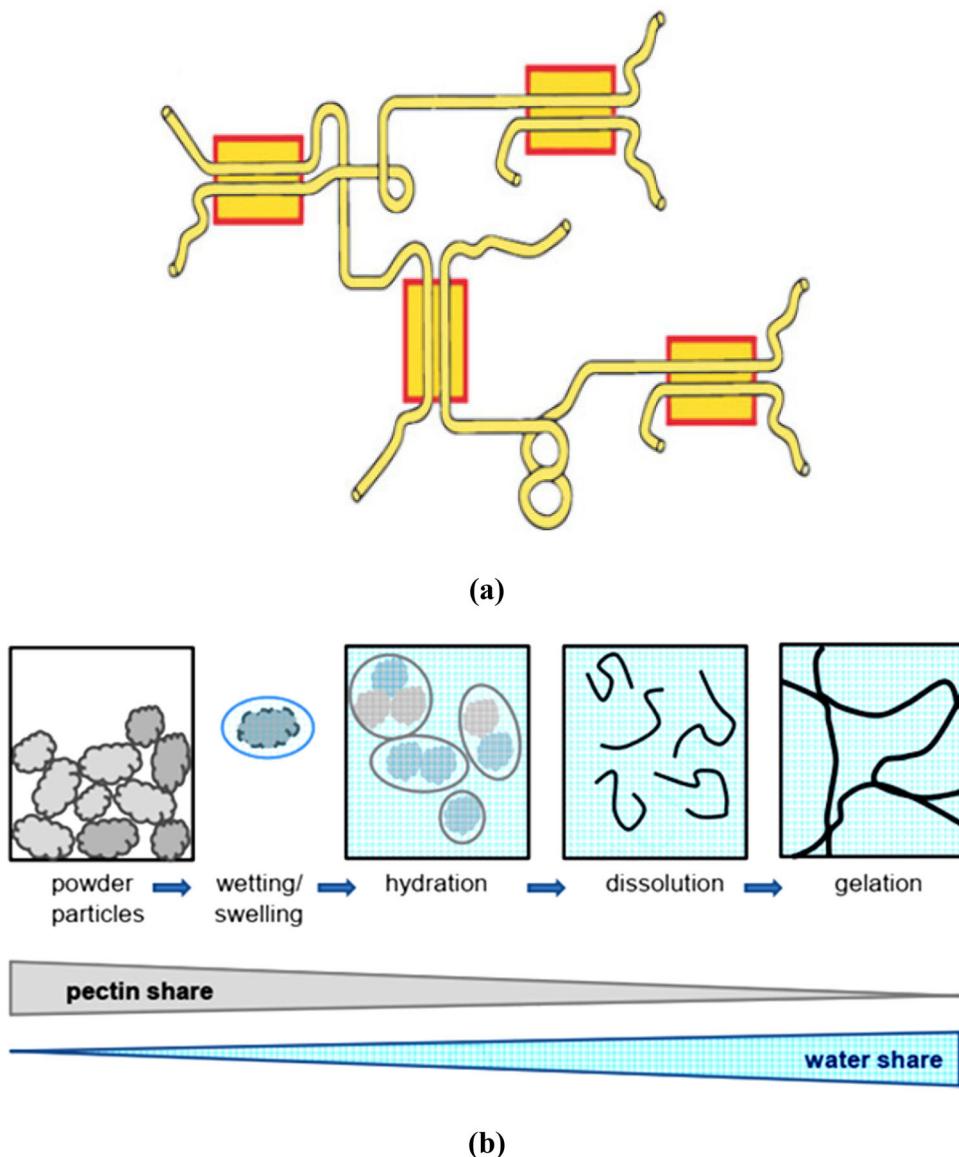
Edible food-ink; fat replacer; high-methoxyl pectin; hydrogel; low-methoxyl pectin; oleogel

## Introduction

“Hydrogels” are polymeric materials that can absorb and retain water in their three-dimensional (3D) network (Ahmed 2015). Due to their unique properties, the applications of hydrogels span across a broad range of fields such as food processing, cosmetics, drug delivery, tissue engineering, flexible electronics, soft machines and robotics, sensors and actuators, thermal insulation and so on (Cao and Mezzenga 2020). In the food industry, hydrogels are predominantly used as thickeners and stabilizers. For food applications, the hydrogels must be derived from plant-based biopolymers such as polysaccharides and proteins. Indeed, the concept of food gels became prominent when Henry Braconnot, a French chemist, elucidated the gelation capability of a biopolymer, namely, pectin, in 1825. He coined the term “pectin” to signify a colloidal substance that he found responsible for the gelation of a heated mixture of fruits, water, and sugar upon cooling. This discovery marked the beginning of a versatile and soft polymer – “pectin hydrogel.”

Pectin is a pectic polysaccharide-based biopolymer derived from the primary cell wall and the intracellular layer of higher plants (Van Buren 1991). It is composed of  $\alpha$ -(1→4)-linked polygalacturonic acid backbone, intruded by

rhamnose residues and modified with neutral sugar side chains. Carboxyl groups of the galacturonic acid residues of pectin may be esterified with non-sugar components such as methyl or acetyl groups. Depending on the degree of methyl-esterification or methylation (DM), pectin is classified as either low-methoxyl (LMP; DM <50%) or high-methoxyl (HMP; DM >50%) type (Ni and Yates 2002). Across the years, the food industry uses the gelling, thickening and emulsifying capabilities of pectin in products such as jams, jellies, and marmalades. The encouraging findings on pectin’s structure-function relationship in the yesteryears (Willats, Knox, and Mikkelsen 2006) instigated inclusive investigations on formulating and tailoring pectin hydrogels for customized applications. The outcomes of these studies have shown that the scope of pectin gels is beyond just jams and jellies, and interesting applications can be achieved by tailoring its structure and composition. The widespread applications of pectin hydrogels can be attributed to their soft and flexible nature, high water content, intrinsic biocompatibility, unique structure, and resemblance to natural materials (Caló and Khutoryanskiy 2015). Further, pectin gels outweigh the gels derived from other gel-forming biopolymers such as gelatin in their rapid gelation, higher



**Figure 1.** Mechanism of pectin gelation: (a) schematic illustration of a gel network with junction zones (Reproduced with permission from Herbstreith and Fox Corporate Group; [www.herbstreith-fox.de](http://www.herbstreith-fox.de)); (b) schematic representation of the sequence of events during pectin gelation (Reproduced with permission from Einhorn-Stoll 2018).

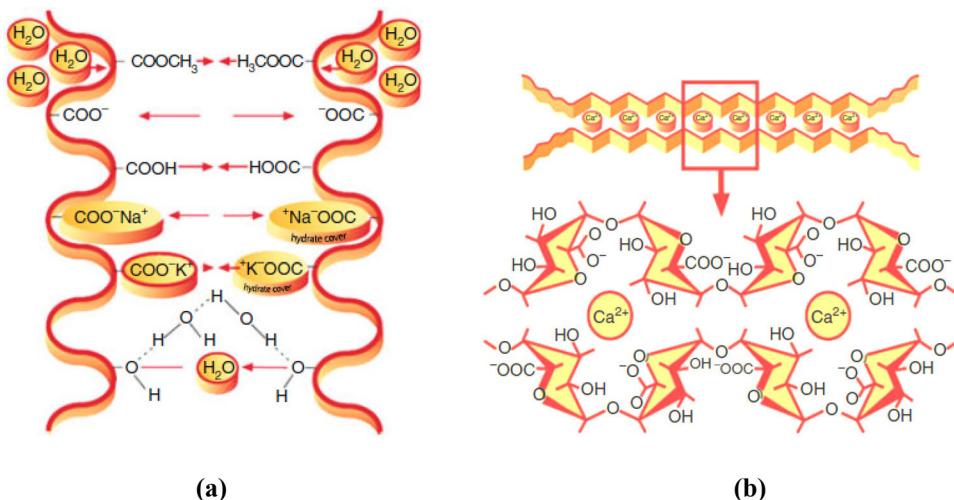
melting point, superior carrier properties for flavoring materials, and fat barrier property (Schrieber and Gareis 2007).

Here, we review the gelling mechanisms of pectin and an array of recently evolved applications of pectin hydrogels. Under the headings of food ingredients/additives, food packaging, bioactive delivery and health management applications, the corresponding sections of this review would explain with pertinent case-studies, the role of pectin hydrogel as a (1) fat replacer; (2) texturizer; (3) edible ink for 3D food printing; (4) packaging film; (5) encapsulant or carrier material for the targeted delivery of nutrients/bioactives, and as a (6) satiety inducer for obesity management. This review intends to provide a comprehensive understanding of the underlying mechanisms by which pectin hydrogels demonstrate each of the aforesaid applications. Further, the physicochemical and structural modifications that enhance pectin hydrogel's suitability for these functions would also be emphasized. A brief overview will also be provided on the recent research trends pertaining to pectin hydrogel,

including the concepts of smart gels, injectable gels, oleogels, aerogels, and xerogels.

### Gelling mechanisms of pectin hydrogels

Hydrogels are formed by linking the neighboring chains of pectin molecules via junction zones to result in a filamentous network of macromolecules. A junction zone (Figure 1a) is constituted by segments from two or more polymer molecules that are held together and stabilized by hydrogen bonding between the carboxyl and secondary alcohol groups and hydrophobic interactions between methyl esters (Sharma et al. 2006; Walkinshaw and Arnott 1981). A substantial amount of water is immobilized in the voids of the resultant network. For instance, at a concentration of <1% in the gels, pectin can immobilize up to 50% of water. Gelation of pectin occurs through a sequence of events (Figure 1b). It commences once the particles of pectin powder



**Figure 2.** Gelling mechanisms of pectin: (a) high-methoxyl pectin; (b) egg-box model of low-methoxyl pectin (Reproduced with permission from Herbstreith and Fox Corporate Group; [www.herbstreith-fox.de](http://www.herbstreith-fox.de)).

are wetted or swollen in water. Subsequently, the pectin particles hydrate and dissolve in water, often under heating or cooking conditions. Then, depending on whether the pectin is of high-methoxyl or low-methoxyl type, the gel is formed by following either the cold gelation or ionotropic gelation mechanism, respectively (Einhorn-Stoll 2018).

### Cold gelation mechanism of high-methoxyl pectin

Cold gelation of HMP is a 2-step process (Burey et al. 2008). The first step involves temperatures higher than 50 °C to achieve the suspension and dissolution of pectin in water. During heating, hydrophobic interactions are formed between the non-polar methoxyl/ester groups in the polymer chain of pectin. The junction zones are stabilized by these hydrophobic interactions combined with the hydrogen bonds (Figure 2a). The hydrophobic effects commence from the unfavorable interactions between water molecules and the methoxyl groups of pectin molecules. Consequently, the methoxyl groups stimulate changes in water structure and thereby reduce its entropy (Thakur, Singh, and Handa 1997). Owing to their incompatibility with water, the methoxyl groups are forced to coalesce and reduce their area of interface with water to lessen the effects of the above entropy change. Actual gelation occurs during the subsequent cooling step, wherein the hydrophobic interactions are loosened, cleaved, and then replaced by hydrogen bonds between hydrophilic carboxyl (-COOH) groups on the pectin chain and also between the hydroxyl (-OH) groups of neighboring molecules (Oakenfull and Scott 1984).

The critical pre-requisites for the cold gelation mechanism of HMP are low pH (i.e., lower than the pKa of pectin which is typically in the range of 3.5–4.5 [Sperber et al. 2009]; usually, pH in the range of 2.0–3.0 is used) and low water activity (water activity or  $a_w$  is the ratio between the vapor pressure of water in a substance [P] to the vapor pressure of pure water [ $P_0$ ] at the same temperature). While organic acids are employed to control the pH, soluble solids, mainly sugar (sucrose), is used as the co-solute to reduce

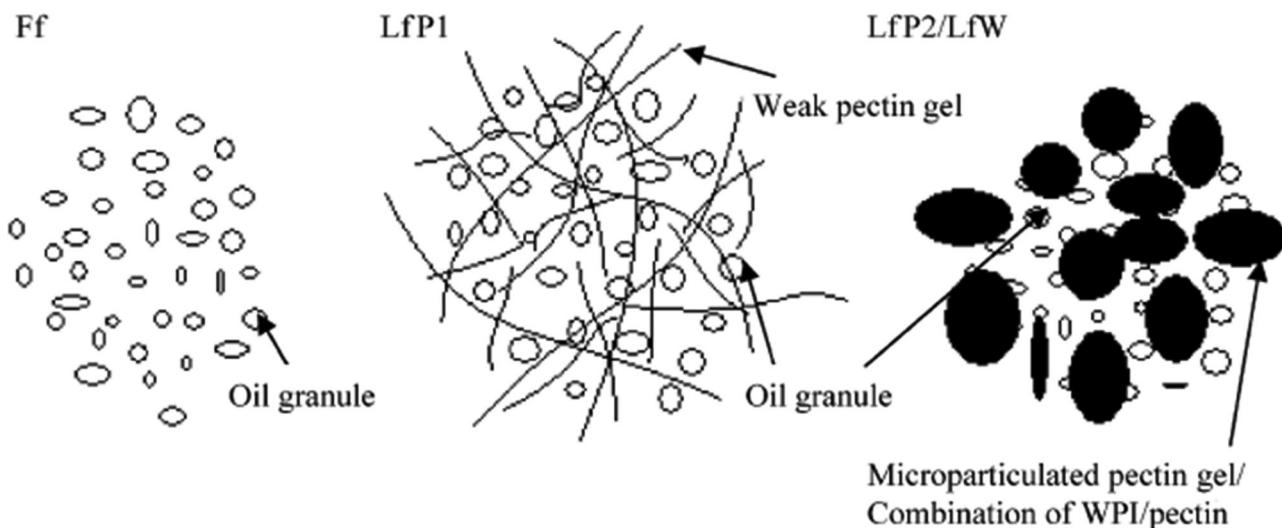
the water activity. The other co-solutes include sorbitol, ethylene glycol, ethanol, and t-butanol.

A low-acidity environment favors the formation of a substantial number of undissociated carboxyl groups (-COOH) and makes them available for hydrogen bond formation (Kastner et al. 2014). Before adding acids to the aqueous pectin dispersion, which may be neutral or only slightly acidic, the unesterified carboxyl groups are present as partially ionized salts. With the addition of acids, the carboxyl ions are converted to unionized carboxylic acid groups. Consequently, the number of negative charges is reduced, which lowers the attraction between pectin and water molecules, besides lowering the repulsive forces between pectin molecules. The above events reduce the dispersibility of pectin. On cooling, this unstable dispersion of less-hydrated pectin forms a gel, which is a continuous network of pectin holding the aqueous solution (Raj et al. 2012).

On the other hand, sugar at a level of >50% (55%–75%) reduces water activity and stabilizes the junction zone by promoting hydrophobic interactions. Sugar reduces the hydration capacity of pectin by competing for the available water. Due to their hydrophilicity, sugars bind a significant amount of available water in the system, increasing the proximity between pectin macromolecules and facilitating the bond formation (Evageliou, Richardson, and Morris 2000; Fraeye et al. 2010; Kastner, Einhorn-Stoll, and Senge 2012). The above sequence of events reduces the ability of pectin to remain dispersed. Eventually, cooling this unstable dispersion of less hydrated pectin results in gel formation, which is a 2D network of pectin molecules in which water, sugar, and acid are immobilized. Further, the immobilization of water in the pectin gel network relies upon the primary inclusion of water in the network and the ability to prevent or, in any case, delay the water retention from the gel structure. Generally, HMP gels are thermally reversible and soluble in hot water (Raj et al. 2012).

### Ionotropic gelation mechanism of low-methoxyl pectin

Low-methoxyl pectin (LMP) forms gel via a single-step process known as the “ionotropic gelation,” which is



**Figure 3.** Pectin hydrogel as fat replacer: schematic representation of the hypothesis on the interaction between fat mimetic and oil-in-water emulsion (Reproduced with permission from Liu, Xu, and Guo 2007).

independent of temperature, sugar, and acid. Typically, this mechanism involves the cation-mediated gelation (mainly,  $\text{Ca}^{2+}$ ) of negatively charged carboxyl groups ( $-\text{COO}^-$ ) available on the backbone of LMP (Glicksman 1983). The divalent metal cations promote interactions between polyanion-cation-polyanion pairs of carboxylate groups on the adjacent helices of pectin chains (Patel, Srinatha, et al. 2014). The above phenomenon has been described as “ionic molecular association,” and the gelling mechanism of LMP is well-known as the “egg box” model (Figure 2b) (Grant et al. 1973). The name “egg box” signifies the structure of the junction zone formed by the calcium-induced association between chains of homogalacturonides. During the initial stages of LMP gelation, two homogalacturonide chains form a dimer in a  $2_1$  symmetry through the calcium ( $\text{Ca}^{2+}$  ions) bridging of parallel facing chains (Morris et al. 1982). An effective calcium bridging is made possible by the relative rigidity of homogalacturonide chains (Axelos and Thibault 1991).

The formation of calcium bridges requires a certain amount of dissociated carboxyl groups ( $-\text{COO}^-$ ), for which reason, pH during gelation of LMP must be greater than the  $p\text{K}_a$  of pectin. After two pectin chains bind the first  $\text{Ca}^{2+}$  ion and align in an anti-parallel orientation with respect to each other, they promote an easier binding of the next calcium ion followed by a sequence of binding events. The initial dimer association is strongly stabilized by Vander Waals and hydrogen bonding, in addition to electrostatic interactions (Braccini and Perez 2001). Apart from the cold gelation and ionotropic gelation mechanisms, there are other pectin gelation methods, such as physical and chemical cross-linking, which would be explained in the upcoming relevant sections.

and Guo 2007). Consequently, the market demand for low-fat foods is on the rise, and the manufacturers are continually looking for novel ingredients that can replace fat in foods. Primarily, an ingredient that may be incorporated in food products to wholly or partially replace fat, more specifically triglycerides, is known as a “fat replacer” or “fat mimetic.” Studies have demonstrated the function of pectin microgel as a fat replacer in foodstuffs. According to early studies, only ionically bound, i.e., calcium-gelled pectin, was considered suitable for use as fat replacers. Later, it was proved that covalently cross-linked pectin could also function as a fat replacer and as an emulsifier (Søndergaard, Juul, and Nørbøge 2000).

The role of pectin gel as a fat replacer depends on its ability to mimic the mouthfeel of fats by contributing a higher viscosity to the liquid phase in the mouth (Swanson 2003). The extent of an increase in product viscosity depends on the tangly (twisted or intertwined) interaction between the 3D structure of pectin gel and oil granules (Figure 3). Another factor that influences the fat replacement function is the particle size of fat mimetics. To be a better fat replacer, the particle size of the pectin gel must be similar to that of the oil granules, which necessitated the development of pectin microgels using an appropriate size reduction method. The microgel particles are identical to fat particles with respect to their softness and deformability and hence can mimic the physical and sensory characteristics of emulsified fat (Worthy 1991).

LMP was used as a fat mimetic in mayonnaise at a replacement level of 50%. Pectin gel in three different forms was used for this purpose. First, a microparticulate pectin gel (LfP1) was prepared by dissolving pectin in water in the presence of calcium, followed by chopping the gel to a defined mass and shearing the coarse particles to form non-spheroidally shaped gel particles. The other two formulations were a weak pectin gel (LfP2) and a microparticulated gel of whey protein isolate and LMP (LfW). The low-fat mayonnaises prepared using the above pectin formulations had significantly lower calories but higher water content than the full-fat product. On increasing, maintaining and decreasing the shear rate sequentially, it was found that all

## Food ingredient/additive applications of pectin hydrogel

### Pectin gel as a fat replacer

Due to the reported health concerns, food manufacturers are in an urge to reduce the amount of fat in the diet (Liu, Xu,

the mayonnaise samples exhibited thixotropic and shear thinning behavior. However, the thixotropy of high-fat mayonnaise was higher by 9-fold, 1.6-fold and 4-fold than LfP1, LfP2, and LfW, respectively. The lower thixotropicity of low-fat samples was attributed to the progressive breakdown of the product's structure with an increase in the shearing time (Abu-Jdayil 2003).

The tangled (twisted) interaction between the 3D structure of pectin gel and oil granules resulted in greater particle size as the samples were scarcely dispersed in solution. The compact packing of oil droplets into the network of weak pectin gel was hypothesized to be responsible for the strengthened network structure, solid-like properties and deformation resistance of the mayonnaise. Thus, it was evident that compared to other low-fat samples, the structure of mayonnaise prepared with weak pectin gel was more similar to that of full-fat mayonnaise. Also, it had a better fat mimetic activity and resulted in low-fat mayonnaise with lower-calorie but similar texture as the full-fat mayonnaise (Liu, Xu, and Guo 2007).

In another study, oil-filled hydrogel particles were prepared by controlling the phase separation of a biopolymer mixture, which are primarily double emulsion systems (oil-in-water-in-water or O/W/W) containing fat droplets (0%–1%), sodium caseinate (1.5%–3%) and high-methoxyl pectin (1.5%–3%) at pH 5. In a simple method, all the components were mixed at pH 7, followed by reducing the pH of the biopolymer mixture to 5. pH reduction facilitated the electrostatic attraction-mediated adsorption of pectin to the surfaces of caseinate-rich hydrogel particles. At pH greater than the isoelectric point (pI), negatively charged polysaccharides can bind to proteins due to electrostatic attraction between anionic groups on the polysaccharides and cationic patches on the protein surfaces (Kayitmazer et al. 2013). Consequently, the resultant oil-filled hydrogel particles comprised of fat droplets were entrapped within caseinate-rich hydrogel particles, which in turn were dispersed inside a protective layer of anionic pectin-rich phase. They were found to have a spheroid shape with a mean particle diameter of  $\sim 10\text{ }\mu\text{m}$ . The oil-filled hydrogel particles enhanced the lightness and viscosity of aqueous solutions and hence were proposed to replace fat droplets in reduced-calorie products. The mechanism of fat replacement by the filled hydrogel particles was hypothesized as due to the presence of a larger number of free pectin molecules in the aqueous phase, more non-spherical particles, or less densely packed particles (Chung et al. 2013).

### **Pectin hydrogel as a food texturizer**

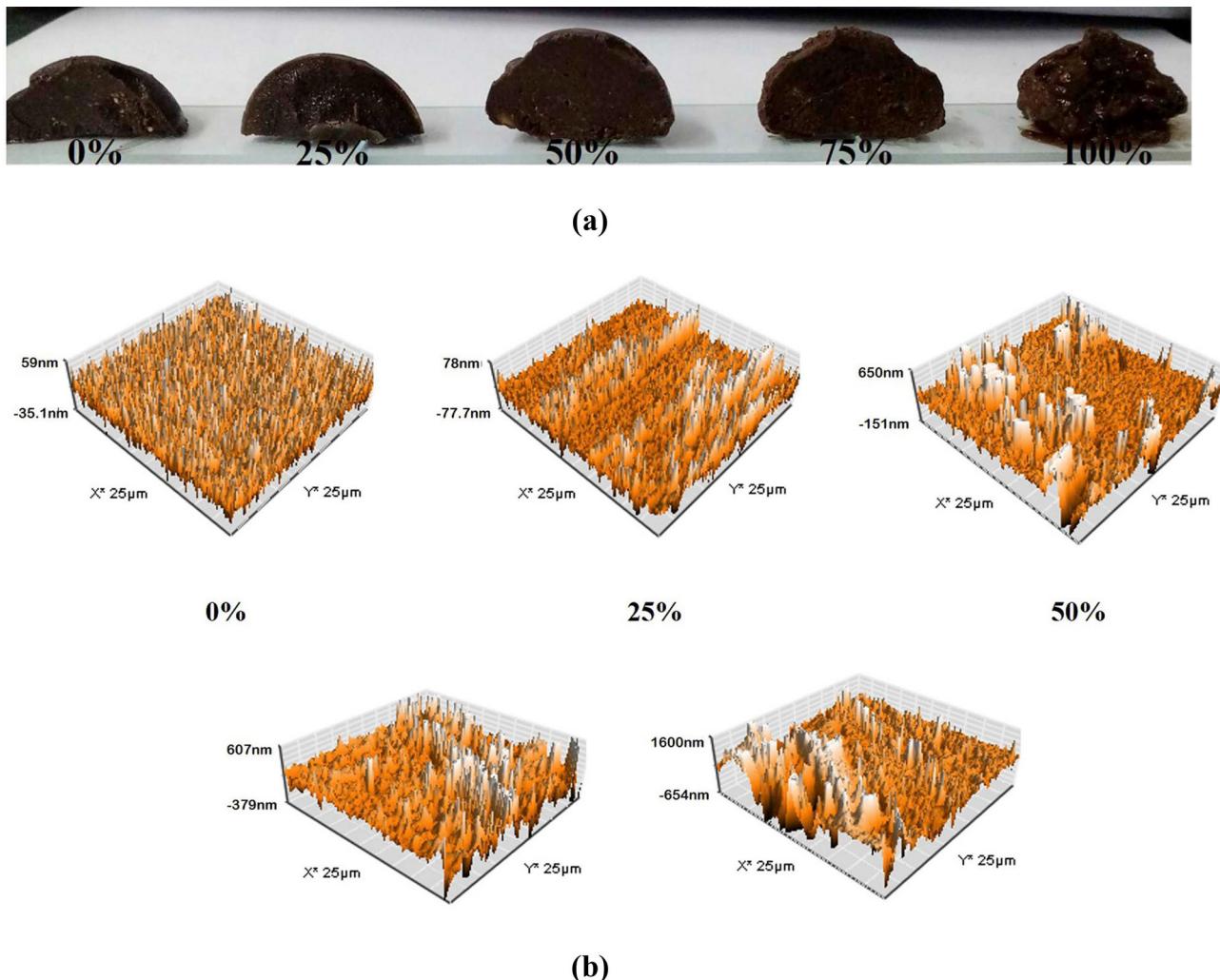
Granulated starch is the widely used texture modifier, thickening agent and fat replacer in the food industry. Although starch granules are effective in the above roles, excessive levels of digestible starch may lead to diabetics. Various investigators have reported pectin's efficacy as a texture modifier and its ability to replace starch granules. Pectin (0%–0.2% wt/wt) and gelatin (0.5% wt/wt) were mixed at pH higher than the isoelectric point of gelatin (pH 9, 30 °C) to prepare

hydrogel microspheres. Subsequently, when the pH was reduced to 5, the above mixture spontaneously formed micron-sized particles due to the electrostatic attraction between cationic gelatin and anionic pectin accomplished via complex coacervation. The major challenge was to tailor the particle size of hydrogel particles to match that of starch granules.

The ratio of gelatin-to-pectin is a vital determinant of the biopolymer charge stoichiometry and intensity of electrostatic interactions, which affects the formation and properties of hydrogel particles (Kizilay, Kayitmazer, and Dubin 2011; Schmitt et al. 1999). The proportions of pectin and gelatin for the preparation of hydrogel particles were fixed based on the following criteria: (1) to ensure that the resultant hydrogel microspheres were true coacervates between gelatin and pectin and not due to sole gelation of gelatin molecules (WHO 2013); (2) to obtain a final mixture in a fluid form rather than gelled form and (3) to match the dimensions and structure of the starch granules. On the other hand, pectin concentration was a key factor influencing the particle size distribution and mean particle diameter of the coacervates formed. At 0.1% (wt/wt) pectin, particles in the mixture were found to coalesce. But, at 0.01% (wt/wt) pectin, the mixture contained individual, translucent, and spherical particles of similar microstructure and size as swollen starch granules ( $D_{3,2} \approx 23\text{ }\mu\text{m}$ ). Therefore, the coacervates were prepared by mixing 0.01% (wt/wt) pectin and 0.5% (wt/wt) gelatin to mimic the size and microstructure of starch granules.

The pellet/sediment obtained after centrifugation of the above suspension constituted the coacervate phase containing the hydrogel particles. The appearance of the coacervate phase was similar to that of starch paste. The coacervate phase was composed of closely packed spherical hydrogel particles, the morphology of which was the same as that of starch granules packed within a starch paste. The flow curve profile (shear viscosity versus shear rate) of the coacervate phase revealed a shear-thinning behavior (power-law index,  $n = 0.7$ ), similar to that of starch paste. The coacervate phase had a substantially higher yield stress ( $\sim 300\text{ Pa}$ ) than the starch paste ( $\sim 14\text{ Pa}$ ), which was due to the jamming phenomenon during which the coacervate phase contained closely packed hydrogel particles. Unlike the swollen starch granules, the hydrogel particles appeared less deformed and retained their spherical shape under the jammed conditions. These hydrogel particles may be used as texture modifiers in reduced-calorie foods, with an added advantage of controlling obesity and diabetes. By adjusting the effective concentration of hydrogel particles, comparable textural attributes (yield stress and shear viscosity) as those provided by starch granules can be obtained (Wu, Degner, and McClements 2014).

Similarly, pectin + sodium alginate hydrogels were prepared using citric acid as a cross-linking agent to replace the cocoa butter and increase the thermal stability of chocolates. Among the chocolates containing different levels of hydrogel incorporation, that incorporated with 50% (vol/vol) of hydrogel showed enhanced glossiness (Figure 4a). Atomic force microscopy (AFM) showed lesser surface roughness of chocolates with 50% (vol/vol) of hydrogel. Increasing the



**Figure 4.** Chocolates containing pectin hydrogels as texturizer: (a) photographs of chocolates containing pectin hydrogel at different levels; (b) AFM images representing the surface roughness of hydrogel dispersed chocolates (scan size:  $25 \times 25 \mu\text{m}$ ) (Reproduced with permission from Francis and Chidambaram 2019).

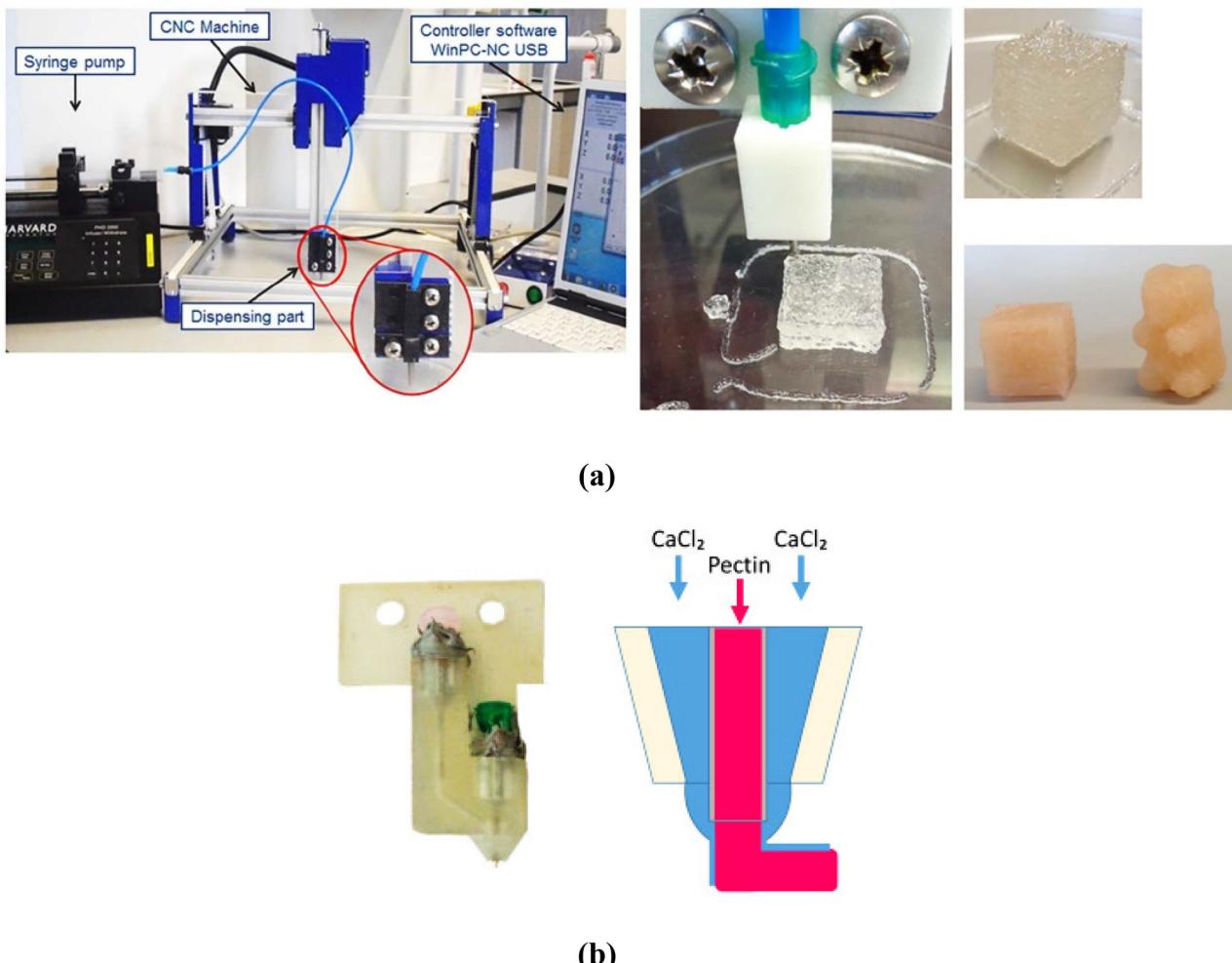
percentage of hydrogel dispersion in chocolate increased the cone size and concentration in the 3D projections and thereby the surface roughness (Figure 4b). Also, the above said chocolate product exhibited the required form-V cocoa butter polymorphism ( $\beta$  polymorph), highest melting resistance ( $80^\circ\text{C}$ ) and non-Newtonian rheology explained by the Casson model, which is the usual model that describes the flow behavior of chocolates (Francis and Chidambaram 2019). Similarly, in cakes, pectin hydrogels were found to replace the shortenings effectively (Lim, Ko, and Lee 2014).

#### Pectin gel as an edible ink for 3D food printing

3D food printing is an “additive” manufacturing (AM) process involving precise layering of tiny semi-liquefied food particles on top of each other. It is an innovative food structuring technique to develop personalized processed foods with unique shapes, structure, texture, and flavors (Anandharamakrishnan and Ishwarya 2019). Scarce availability of printable food materials is the shortcoming that limits the commercialization of 3D food printing. The printability of a material is determined by its viscosity, consistency, and solidifying properties. Based on printability, food materials can be classified as natively

printable and non-printable. While natively printable materials can be smoothly extruded from a syringe (Cohen et al. 2009), non-printable materials are not innately printable. Still, they can be made printable by adding food-grade hydrocolloids (Sun et al. 2015).

In the search for novel printable food materials, LMP gel has been identified as a potential food ink for 3D printing applications. But, LMP gel is not always suitable for 3D printing as it lacks the iron or calcium that provides the required viscosity. However, it can be rendered printable using additives such as sugar sirup, bovine serum albumin (BSA), and calcium chloride ( $\text{CaCl}_2$ ). Although LMP gelation does not require sugar, the addition of sugar sirup enhances the viscoelastic properties of the pectin gel (Grosso and Rao 1998; Fu and Rao 2001; Lopes da Silva and Rao 2006) and provides sweetness. The concentration of pectin and sugar influences the viscosity of food-ink and thereby the build quality of printed objects. Also, pectin concentration determines the firmness and strength of the 3D printed product. The pectin concentration can be chosen according to the target texture. For instance, 15 g/L pectin was found sufficient to obtain a soft-textured product with  $G'$  in the range of 0–60 kPa. A higher concentration of pectin at 25 g/



**Figure 5.** Pectin gel as edible ink for 3D food printing: (a) illustration of the 3D printer setup and process for printing gummy bear-shaped candies (Reproduced with permission from Vancauwenbergh et al. 2017); (b) picture of the coaxial print-head and schematic of the junction between inner flow and outer flow (Reproduced with permission from Vancauwenbergh et al. 2018).

$L$  should be used to obtain hard-textured products with their  $G'$  varying between 60 kPa and 100 kPa. For much harder products ( $G' > 100$  kPa), a still higher concentration of pectin at 35 g/L must be chosen (Vancauwenbergh et al. 2017). To acquire suitable flowability of the gel, CaCl<sub>2</sub> is added to the LMP food-ink according to the following stoichiometric ratio:

$$R = \frac{2[Ca^{2+}]}{[COO^-]} \quad (1)$$

A partial cross-linking of the gels during printing is said to be attained when the  $R$ -value falls in the range of 0.2–0.5. While a high  $R$ -value impacts the printability conditions and properties of the 3D printed objects, a low  $R$ -value due to insufficient calcium may lead to partial spreading during printing and swelling during the post-treatment (Choi et al. 2015). LMP/Ca<sup>2+</sup> gel showed highly structured and viscoelastic behavior ( $G' > G''$ ) (Choi et al. 2015), due to which it did not spread during printing, and the 3D shape of the object was sustained with time. Irrespective of the pectin concentration, calcium ions significantly influenced the textural properties of printed pectin objects. Therefore, a hypothesis was proposed that the mechanical properties of LM-pectin gel may be proportional to its cross-link density

(Fraeye et al. 2010). The hypothesis mentioned above is in concurrence with the texturizer property of pectin gels, as discussed earlier.

The texture of LMP gels can be altered by just varying the amount of calcium used for cross-linking. While low calcium at high pectin content leads to an elastic gel, high calcium with low pectin content produces a more brittle product, probably with a certain degree of syneresis (Vancauwenbergh et al. 2018). Especially, for the optimum texture of 3D printed fruit and vegetable-based products, pectin-calcium interactions hold relevance as the cross-linked pectin in the cell wall offers cell-cell bonding and mechanical strength of tissues (Fraeye et al. 2010).

A prototype 3D printer was developed by the KU Leuven University in Belgium, which employed LMP gel as the food ink to prepare candy of different shapes (Figure 5a) (Vancauwenbergh et al. 2017). Candy is an example of water-based porous simulants, and 3D printing of candy was perceived as a new approach to print 3D cellular plant tissues for innovative food manufacturing. The amount of sugar that can be added without changing the printability and extrudability of the product at room temperature is crucial. By varying the stirring rate (3200, 6700, 10200 rpm) and the concentrations of LMP (15, 35, 55 g/L), CaCl<sub>2</sub> (12.5,

15, 17.5 mM), BSA (2.5, 5 g/L), and sugar sirup (0, 25, 50% [vol/vol]), pectin gels with tunable microstructure and texture properties were successfully 3D printed. After printing, the printed object was incubated in a 300 mM solution of  $\text{CaCl}_2$  for 10 min to achieve complete gelation. The printed objects were then manually sprayed with 300 mM calcium solution every 10 min for 90 min. This was done to solidify the pectin gel and allow the calcium ions to diffuse through the gel. The spraying time was increased according to the sample size (Vancauwenbergh et al. 2018).

As an advancement over the previous study, subsequent studies by the same authors focused on designing a coaxial extrusion print-head (Figure 5b) for the 3D printing of pectin-based food simulants (Vancauwenbergh et al. 2018). Coaxial print-head has an accurate control over the gelation and textural properties of the pectin object. The objects printed by coaxial and simple extrusion methods had similar Young's moduli, but their volume and final  $\text{Ca}^{2+}$  concentration were considerably influenced by the method of inducing gelation. While the objects printed by simple extrusion can be considered continuous bulk material, those printed by coaxial extrusion could be visualized as a stack of layers. The pectin gelation occurring during printing eliminates the need for the post-printing incubation of printed objects in  $\text{CaCl}_2$ . This is an important advantage of extrusion-based 3D printing in reducing the manufacturing time of an object.

Pectin gels have also been used to improve the structural stability of 3D edible cubes prepared from some bio-inks. For instance, an 11% pectin solution provided the required viscosity to a banana-based paste to make a nutritionally customized 3D-printed snack for children of 3–10 years old. The pectin solution was added at a mass fraction of 30% to a food-ink formulation, in which the remaining 70% was banana, white canned beans, dried nonfat milk, lemon juice, dried mushrooms (*B. edulis*), and ascorbic acid. The pectin addition prevented the phase separation between water and vegetable tissue during the deposition (Derossi et al. 2018). Table 1 presents the findings of recent investigations that employed pectin hydrogel as edible ink for 3D food printing.

## Role of pectin hydrogel in food packaging

Active investigations are underway on finding novel and sustainable packaging films to replace non-biodegradable petroleum-based food packages. Nowadays, food packaging materials are expected to have antibacterial and antioxidant properties to prolong the shelf-life of food products. Their proven biodegradability and flexibility confer interesting food packaging applications upon the pectin gels. Besides, pectin-gel-based packaging materials exhibit good oxygen barrier properties, besides reducing the respiration rate and oxidation of foods (Mafsoonazad and Ramaswamy 2008). These films also possess good hardness and adhesiveness.

Despite the above merits, the performance of pectin gel-based films in food packaging is affected by their rigidity, brittleness and poor water barrier properties. The above limitation can be overcome by preparing a composite film of pectin combined with one or more renewable and

environment-friendly biopolymers. Some biopolymers often used in combination with pectin include cellulose (Ye et al. 2019), aloe vera gel (López-Mata et al. 2018), sodium alginate (Solak and Dyankova 2014), and carrageenan (Alves et al. 2011). Properties of the composite film are dependent on the nature of constituent biopolymers. Reactivity between the functional groups of constituent polymers in the composite film results in unique and novel structures with improved properties (Farris et al. 2009).

Pectin-cellulose (PC) composite films were prepared by incorporating natural tea polyphenols (TP) and cinnamaldehyde (CA) to confer them with antioxidant and antibacterial properties, respectively (Figure 6a). The scanning electron micrographs (Figure 6b) revealed that the pectin-cellulose composite films with and without the addition of antioxidant and antimicrobial agents exhibited similar dense surface morphologies without any pores. Thus, the addition of TP and CA did not cause any significant modification in the surface morphology of the pectin-cellulose composite film. But, TP and CA increased the roughness and reduced the thermal stability of pectin-cellulose films. The PC composite films without TP and CA did not have any antibacterial or antifungal activity, indicated by the absence of inhibition zones against *E. coli*, *C. albicans*, and *S. aureus*. Clear inhibition zones were observed in PC films loaded with tea polyphenols and cinnamaldehyde, which were found to increase with an increase in the concentration of TP and CA. This observation confirmed the excellent bacterial and fungal inhibition activities of PC composite films. Interestingly, pectin composite films (without any TP loading) exhibited an inherent but mild DPPH free radical and hydroxyl radical scavenging activity (33%) (Ye et al. 2019) due to the hydrogen supplied by cellulose and pectin (Luo et al. 2016).

Pectin has also been used in combination with proteins to prepare composite films. Mixing pectin with proteins produces complexes suitable for making gels in the form of sheets, membranes, and coatings (Farris et al. 2009). Composite hydrogels were prepared from gelatin and LMP (DE = 7) by leveraging their charge and functional properties to produce films with improved properties. In the first step of the process, a reversible physical hydrogel or polyion complex was formed by the ionic interactions between positively charged gelatin and negatively charged pectin through electrostatic forces between  $\text{NH}_3^+$  and  $\text{COO}^-$  (Figure 6c). The homogeneous molecular arrangement of these physical hydrogels resulted in improved mechanical and water resistance and a 10-fold reduction in swelling ability compared to individual polymers. In the second step, glutaraldehyde (0.3%) was added to the polyion complex that chemically cross-linked the gelatin resulting in permanent chemical hydrogels. The addition of glutaraldehyde introduced new bonds between gelatin molecules at the intermolecular level. It led to a heterogeneous microstructure with a distinct web-like conformation in which pectin molecules were present (Figure 6d). After formation, the hydrogels were degassed, cast, and subjected to evaporation in a vacuum oven at 40 °C for 24 hours. The composite films prepared from permanent pectin-gelatin complex hydrogel showed higher tensile strength (by 26%) and elongation at

**Table 1.** Applications of pectin hydrogel in 3D food printing.

S. No.	Source of pectin	Type of pectin	Type of printing	Formulation of pectin ink	Major findings	References
1.	Citrus peel	<ul style="list-style-type: none"> <li>Low methoxyl pectin</li> <li>DE: <math>12\% \pm 0.5\%</math></li> </ul>	Extrusion deposition	<ul style="list-style-type: none"> <li>Pectin solutions: 30 or 70 g/L LMP with and without 0.2% (wt/vol) BSA</li> <li>CaCl<sub>2</sub> solutions: 26 or 40 mM CaCl<sub>2</sub></li> <li>Post treatment solution: 50 mM CaCl<sub>2</sub></li> <li>Evans blue solution: 0.5% (wt/vol)</li> </ul>	<ul style="list-style-type: none"> <li>Land-plant cells can be encapsulated in pectin gel and can be 3D printed with high accuracy and reproducibility</li> <li>The encapsulation of cells in bio-ink resulted in decreased mechanical and structural properties</li> </ul>	Vancauwenbergh et al. (2019)
2.	Commercial pectin	<ul style="list-style-type: none"> <li>Low methoxyl pectin</li> <li>DE: <math>19\% \pm 0.8\%</math></li> </ul>	Coaxial extrusion	<ul style="list-style-type: none"> <li>15 g/L pectin with 10 mM CaCl<sub>2</sub></li> <li>25 g/L pectin with 12.5 mM CaCl<sub>2</sub></li> </ul>	<ul style="list-style-type: none"> <li>3D printing of pectin based food stimulant was done successfully with a coaxial print-head</li> <li>Coaxial extrusion facilitated the gelation of pectin, without the need for any treatment after printing</li> </ul>	Vancauwenbergh et al. (2018)
3.	Apple	<ul style="list-style-type: none"> <li>Low methoxyl pectin</li> <li>DE: 12%–25%</li> </ul>	Extrusion method	<ul style="list-style-type: none"> <li>Carboxylated-cellulose nanofibrils gel with bleached sulfite pulp (1.19%)</li> <li>Sodium Hydroxide: 98%</li> <li>Hydrochloric acid: 37%</li> </ul>	<ul style="list-style-type: none"> <li>Pectin based bio-ink suitable for 3D printing was prepared</li> <li>Increase in pectin concentration improved the printability</li> <li>Low speed printing was considered a drawback</li> </ul>	Cernencu et al. (2019)
4.	Citrus peel	Low methoxyl pectin	Extrusion	<ul style="list-style-type: none"> <li>Pectin: 15–55 g/L</li> <li>CaCl<sub>2</sub>: 12.5–17.5 mM</li> <li>BSA: 0–5 g/L</li> <li>Sugar sirup: 0–50% (vol/vol)</li> </ul>	<ul style="list-style-type: none"> <li>Considered as the first step to produce printed pectin based porous foods</li> <li>Pectin concentration had a positive impact on strength, firmness and elasticity of the printing ink gel.</li> <li>BSA increased the aeration and stability of the edible ink</li> <li>Final product was a fruit based formula for children of 3–10 years, which contained 5%–10% of the required energy</li> <li>Proved the possibility of personalized food with desired shape and dimension with nutritional value</li> </ul>	Vancauwenbergh et al. (2017)
5.	-NM-	Pectin powder E440	Extrusion	<p>70% of the printable food formula:</p> <ul style="list-style-type: none"> <li>Banana: 73%</li> <li>Canned white beans: 15%</li> <li>Dried nonfat milk: 6%</li> <li>Lemon juice: 3.0%</li> <li>Dried mushroom: 2.0%</li> <li>L-ascorbic acid: 0.5%</li> </ul> <p>30% of the printable food formula:</p> <ul style="list-style-type: none"> <li>Pectin solution: 30%</li> </ul>		Derossi et al. (2018)

-NM: details not mentioned.

break (by 24%) than the pure gelatin films. Cross-linking led to further improvement in the above properties, which was accredited to the electrostatic interactions between the oppositely charged functional groups of pectin and gelatin (Farris et al. 2011).

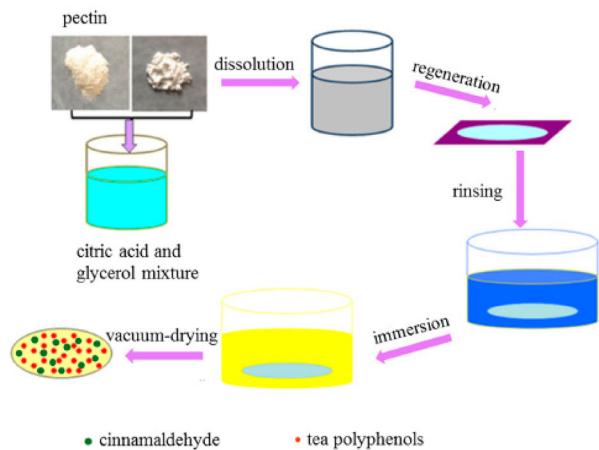
## Applications of pectin hydrogel in bioactive delivery

### Pectin gel for encapsulation and targeted delivery applications

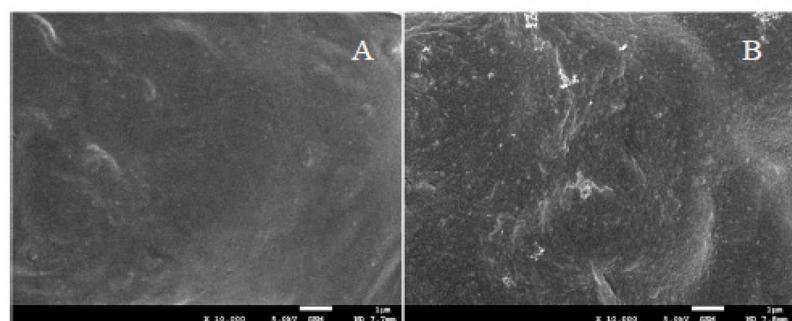
Hydrogel is an interesting choice of wall material for the encapsulation of biomolecules and living cells. It offers a congenial aqueous environment required for their biological functioning of the encapsulated material. Further, its cross-

linked structure protects the encapsulated therapeutic molecules and cells from immune rejection and protease degradation. Cross-linking provides a diffusion barrier that permits the passage of small molecules and solutes within a given size threshold while eliminating any interaction between encapsulated components and the larger molecules involved in immune surveillance and clearance by the humoral and cellular immune responses (ex. complement system and antibodies, macrophages, B- and T-cells) (Figure 7a). The size threshold of allowable molecules can be customized by varying the degree of cross-linking of the hydrogel.

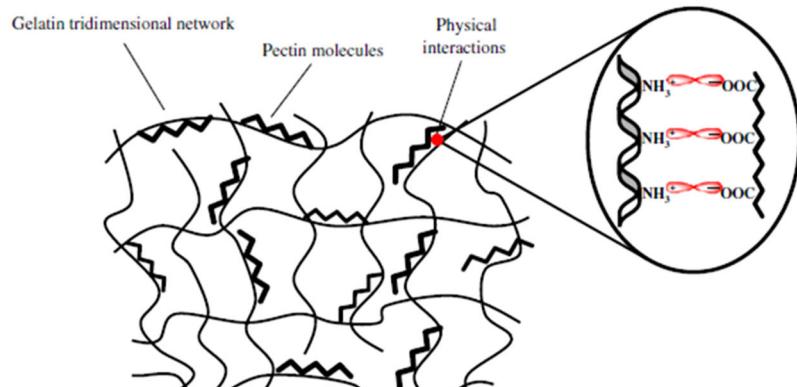
Pectin hydrogel has been found appropriate for controlled release applications. In living cell encapsulation, hydrogels functionalized with biological molecules, say, extracellular matrix proteins or peptides or degradation sequences, can provide the necessary biochemical signals.



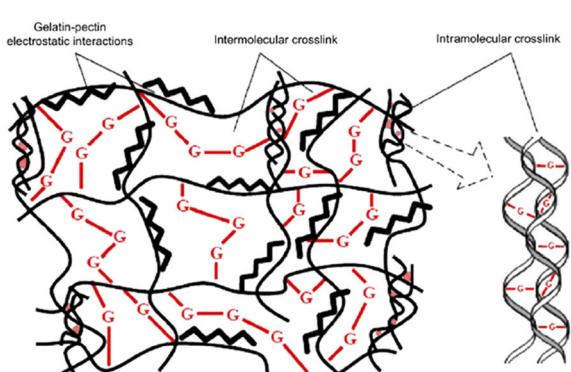
(a)



(b)



(c)



(d)



Also, hydrogels offer a 3D microstructure that can control the mechano-biochemical transduction signals of encapsulated probiotic cells (Pérez-Luna and González-Reynoso 2018). Being a carbohydrate-based polymer, pectin hydrogel is an appropriate choice of wall material to encapsulate food ingredients. The forthcoming sections present the applications of pectin hydrogel for the encapsulation and targeted delivery of bioactive food components.

### **Pectin hydrogels as an encapsulant for probiotic delivery**

Probiotics are live microorganisms that, when consumed in adequate amounts, promote the gastrointestinal health of hosts. *Lactobacillus* and *Bifidobacterium* are the two main bacterial genera that represent the category of probiotic organisms. Encapsulation protects probiotics against the harsh physiological milieu and destructive enzymes in the upper sections of the gastrointestinal tract and preserves their viability during high-temperature processing and storage (Anandharamakrishnan and Ishwarya 2015). Further, the designated delivery system should enable the release of survived probiotics at the target site of action, generally the small or large intestine (Kim et al. 2016). Therefore, an ideal probiotic delivery system should protect probiotics from adverse conditions during fabrication and storage and in the acidic gastric environment so that a sufficient amount of probiotics is available in the site of action.

Pectin hydrogels must possess certain structural features to function as an effective probiotic delivery system. Firstly, pore sizes of the hydrogels must be sufficiently small than the dimensions of bacteria cells to ensure their entrapment in the hydrogel matrix until the breakdown of the network before the release of cells (Cook, Charalampopoulos, and Khutoryanskiy 2014). Thus, the smaller pore size of hydrogels is vital in preventing fast release rates and thus improving the loading capacity. Cross-linking with an inorganic compound such as calcium reduces the permeability of polysaccharide hydrogels. Further, when low methoxyl pectin is employed to prepare gel beads, a low concentration of calcium ( $\text{Ca}^{2+}$ ) ions is preferred to prevent complete gelation and formation of bulk gels. Excess concentration of  $\text{Ca}^{2+}$  ions can lead to competition for interaction with anionic sites ( $\text{COO}^-$ ), leading to weak gels (Sudhakar et al. 2013). In some instances, freezing adjuvants and coating materials are used to prepare pectin hydrogels to provide additional protection to the probiotic cells.

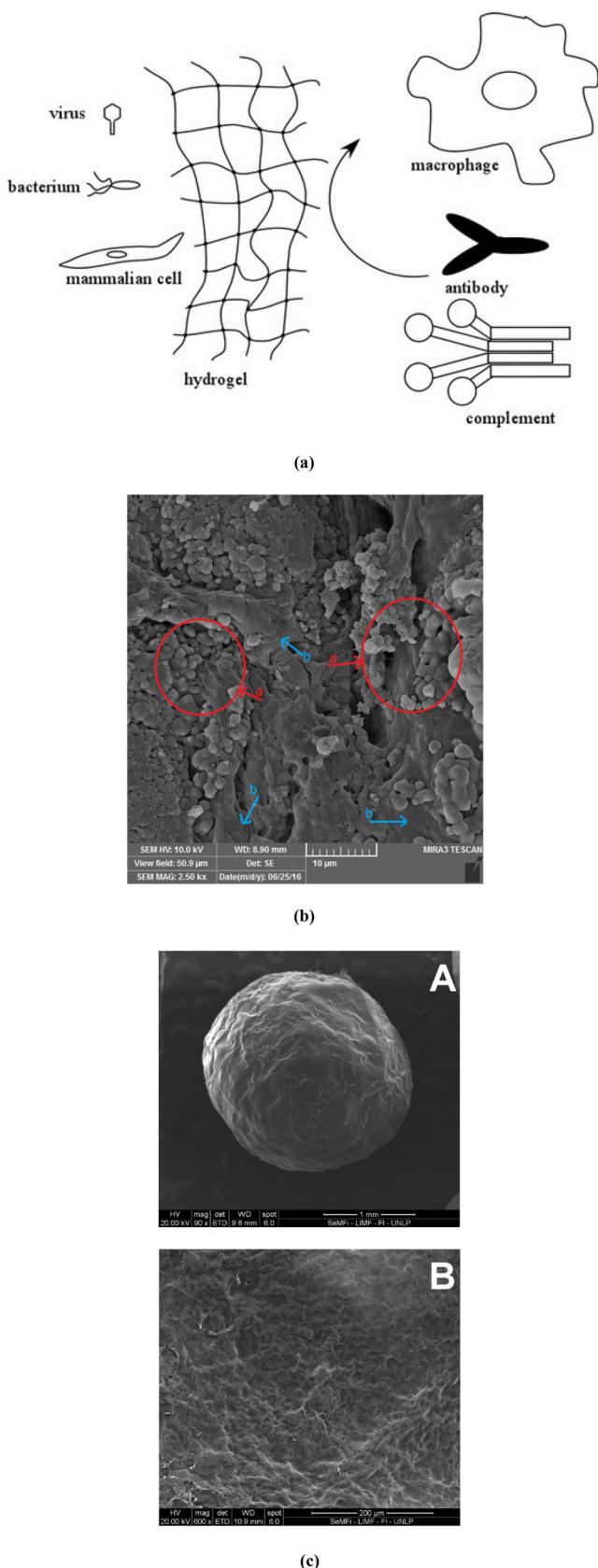
Pectin hydrogel beads containing glucose were employed for the encapsulation of a selected strain of *Lactobacillus rhamnosus*. Probiotics encapsulated within the pectin gel beads were freeze-dried and stored at room temperature for more than one month. Compared to unencapsulated cells, the shelf-life of the encapsulated cells was high during storage under ambient conditions. Glucose acts as a cryoprotectant and

inhibits the formation of ice crystals that can cause cell damage (Pollock et al. 2016). After lyophilization, the pectin gel beads containing probiotic cells were exposed to simulated gastric fluid and simulated colonic fluid containing enzymes. Results demonstrated the protective effect of pectin gel-based encapsulation on the probiotic cells against acidic conditions and enzymes, which was further enhanced by the presence of glucose (Corcoran et al. 2005).

The viability of probiotic cells encapsulated in pectin hydrogel was improved by covering the pectin beads with protective coating materials (Chotiko and Sathivel 2016; Gebara et al. 2013). Accordingly, cells of *Lactobacillus casei* were encapsulated into pectin hydrogel beads, and then the beads were coated with chitosan, a basic polysaccharide. Compared to free cells, the viability of coated and uncoated encapsulated cells was high under the simulated gastric fluid (acidic conditions) and simulated intestinal fluid conditions. Chitosan coating over the pectin hydrogel beads conferred a significantly positive influence on the viability of encapsulated probiotics under simulated gastric conditions (2 h at pH 2.0). Even after two hours of exposure to acidic pH, the viability of probiotics within coated pectin capsules was very high without any significant ( $P > 0.05$ ) reduction in the number of live cells. The cell recovery after two hours was 9.6 logs CFU/mL. However, there was a substantial loss in the viability of cells encapsulated within the uncoated beads to less than 7 logs CFU/mL within one hour. Further, at the end of two hours, the viability dropped to less than 4 logs CFU/mL (Bepelyeva et al. 2017). Thus, it is apparent that the chitosan coating was solely responsible for preserving the cell viability under highly acidic conditions. The positive effect of chitosan coating may be attributed to the electrostatic attractions between the ionized amino groups of chitosan ( $\text{NH}^3+$ ) and the ionized carboxyl acid groups ( $\text{COO}^-$ ) of pectin (Rashidova et al. 2004). In addition, the protective effect of chitosan has also been attributed to its ability to neutralize the  $\text{H}^+$  ions by penetrating through the beads. In other words, chitosan coating functions as a buffering layer to prevent acid ingress (Cook et al. 2013).

Similar effects as above were observed when pectin hydrogel beads containing live cells of *Lactobacillus acidophilus* (Gebara et al. 2013) and *Lactobacillus rhamnosus* (Gerez et al. 2012) were coated with whey protein. The protective effect of whey protein coating was dependent on the electrostatic complexes formed between whey protein and pectin at a pH below the isoelectric point of whey protein (pH 5.2–5.4) (Gentes, St-Gelais, and Turgeon 2010; Santipanichwong et al. 2008). The abovementioned complex formation is facilitated by the interaction between protonated (positively charged) amine groups of whey protein and the negatively charged carboxylic groups of pectin. Subsequently, when the coated pectin hydrogel beads containing probiotics were exposed to intestinal fluids, the coating disintegrated to release the cells (Gebara et al. 2013).

**Figure 6.** Pectin gel as packaging material: (a) schematic representation of the preparation of pectin-cellulose composite films with antioxidant and antibacterial properties; (b) scanning electron micrographs of pectin-cellulose composite films: (A) without the addition of antioxidant and antimicrobial agents and (B) with the addition of antioxidant and antimicrobial agents (Reproduced with permission from Ye et al. 2019); mechanisms for the formation of (c) physical hydrogel formed by charge interaction between pectin and gelatin; (d) pectin-gelatin permanent polyion complex hydrogels (Reproduced with permission from Farris et al. 2009).



**Figure 7.** Applications of pectin gel in encapsulation and controlled delivery: (a) schematic representation of the protective effect of hydrogel on the encapsulated cells (Reproduced with permission from Pérez-Luna and González-Reynoso 2018); (b) scanning electron microscope images of *L. plantarum* cells (indicated by "a" in the figure) encapsulated in pectin/starch hydrogel (indicated by "b" in the figure) (Reproduced with permission from Dafe et al. 2017); (c) SEM micrographs of (A) Surface morphology and (B) Internal structure of iron-pectin beads (Reproduced with permission from Ghibaudo et al. 2018).

Microscopic visualization of the cross-section of encapsulate showed that the probiotic cells were randomly distributed throughout the pectin hydrogel network (Figure 7b). Probiotic cells encapsulated in this hydrogel network must be released into the intestinal fluid to exert their intended health benefits in the human body. The encapsulated cells demonstrated a rapid release profile in the simulated intestinal fluid (SIF at pH 7.4), compared to that in the simulated gastric fluid (SGF at pH 1.2). At the acidic pH of SGF, the anionic carboxylate groups ( $pK_a \sim 4.5$ ) on the pectin backbone were converted into a neutral carboxylic acid. The resultant network in the hydrogen-bonded form counteracted the cell release.

On the other hand, at the alkaline pH of SIF, the carboxylic acid group on pectin backbones was converted to negatively charged carboxylate ions ( $COO^-$ ). The consequent electrostatic repulsion between carboxylate ions inflated the network and enhanced the diffusion of cells from the pectin/starch hydrogels (Pornsak and Nurairat 1998). A complete release of cells was achieved after 8 hours of exposure to the SIF. The release of *L. plantarum* cells from the hydrogel matrix followed the non-Fickian type kinetics (Dafe et al. 2017). Non-Fickian release involves physical processes and thermodynamic forces, i.e., concentration gradient involved in classical Fickian diffusion. In this study, the non-Fickian diffusion involved polymer relaxation in addition to diffusion.

#### Controlled and targeted release of bioactives using pectin hydrogel

Bioactives are a collection of active components that provide enhanced health benefits and facilitate the maintenance of human biological processes (Saura-Calixto 2011). The coacervate or complex formed between LMP and calcium ions has been utilized for the controlled release of organic chemicals, volatiles, or proteins (Liu et al. 2006). Due to their stability in low pH solutions and resistance to enzymatic digestion in the mouth and stomach, calcium pectinate hydrogels have been used to deliver acid-sensitive food bioactives (Sinha and Kumria 2001). Nevertheless, LMP hydrogels are degraded by colon microflora and hence appropriate as a carrier material for controlled release applications (Sriamornsak and Kennedy 2008) and colon-targeted delivery. However, the high swelling behavior of pectin-based gels in physiological environments is a major limitation in using them for colon-specific nutrient delivery.

To tackle the iron deficiency that prevails worldwide, a recent study by Ghibaudo et al. (2018) explored the preparation of iron-pectin beads and evaluated their physiological behavior to demonstrate their prospective usefulness in the food industry. Iron pectin beads were prepared by the ionic gelation method. The resultant beads had a diameter of 1–2 mm and showed high density (1.29 g/mL) and porosity (93.28%), which signified their high permeability even at low pressure. SEM micrographs revealed the spherical shape and smooth external surface of iron-pectin beads (Figure 7c [A]). The internal structure of beads depicted a compact texture with regularly observable holes (Figure 7c [B]). The iron-pectin beads swelled to a greater extent in the

simulated intestinal medium (pH 8) than in the simulated gastric medium (pH 1.25). The above difference in behavior was attributed to the mutual repulsion between negatively charged carboxylic groups ( $-COO^-$ ) (Chang and Lin 2000). At the low pH of the gastric medium, a predominance of  $H^+$  ions results in the protonation of carboxyl groups. This reduced the electrostatic repulsion between polymer chains to render them tighter. Consequently, penetration of liquid into the system was restrained, and solubility of the polymer chains was reduced (Oliveira et al. 2010; Prezotti, Cury, and Evangelista 2014).

On the other hand, at the higher pH of the intestinal medium, carboxyl groups remained in the ionized form, which increased the hydrophilicity to expand the network due to electrostatic repulsion. The conditions mentioned above promoted liquid penetration into the system (Prezotti, Cury, and Evangelista 2014). The presence of hydroxyl anions in the medium facilitated the release of Fe(II), which functioned as cross-linking points of the pectin chains and enhanced water uptake by these systems (Remuñán-López and Bodmeier 1997). As discussed above, the degree of swelling of gel beads is a crucial parameter with respect to the release of an encapsulated substance from its matrix. It depends on the rigidity of gels (Jantrawut, Assifaoui, and Chambin 2013), which in turn is based on the degree of esterification. LMP has the lowest molecular weight (98 kDa), viscosity, gel strength and rigidity compared to other pectins. Finally, the iron released from the pectin beads underwent oxidation, after which it was found as a water-soluble Fe(III)-oligopectin complex. Further, when the digested beads were incubated with Caco-2/TC7 cells for 4 hours, a significant increase was noted in the iron transport relative to the control ( $FeSO_4$ ). Thus, encapsulating iron in pectin beads was an effective approach in overcoming the low efficiency of iron transport and supplementing food products with iron without affecting the sensory properties (Ghibaudo et al. 2018).

In addition to the above studies, various other investigations have used pectin alone or in combination with other hydrocolloids such as alginate, starch, chitosan, and gelatin for the encapsulation of various food bioactives. Table 2 presents a compilation of findings from such studies on the encapsulation applications of pectin hydrogel.

## Health management applications of pectin hydrogel

### Pectin gels as satiety inducers for alleviating obesity

Obesity has been realized as a universal health problem, the incidence of which has nearly increased 3-fold since 1975 (WHO 2018). As the incidence of obesity bears a positive correlation with fat intake, reducing the body's ability to utilize dietary fat has been realized as a significant approach to combat obesity (Bray et al. 2018). In this context, pectin gel has been found to reduce fat absorption in the gut by binding to bile acids and micelle components such as monoglycerides, free fatty acids, and cholesterol (Kaczmarczyk, Miller, and Freund 2012). The exact interaction mechanism between pectin and bile acids is yet to be elucidated

completely. However, it has been found that the interaction between pectin, bile acids and micelle components leads to the binding of water in the chyme. This increases the viscosity, which reduces the diffusion rate of bile acids, which cannot be reabsorbed by the body and thus are excreted (Anttila, Sontag-Strohm, and Salovaara 2008). Similarly, pectin combats obesity by affecting the expression of relevant genes involved in lipid metabolism. In the process of lipogenesis, acetyl-CoA carboxylase is the rate-limiting enzyme, which is regulated by AMP-activated protein kinase (AMPK). Inclusion of dietary fiber such as pectin in the diet has been found to increase the phosphorylation of AMPK, which inhibits the acetyl-CoA carboxylase enzyme (Galisteo et al. 2010).

“Satiety signaling” has been recognized as another means to combat obesity, wherein “satiety” describes the inter-prandial period during which the feeling of fullness (satiation) lingers before hunger returns (Benelam 2009). Based on the studies conducted, it was understood that initially, the pectin gel swells in the acidic environment (pH 1.2) of gastric juice to fill the stomach and thereby induce the satiating effect (Kristensen and Jensen 2011). This is accomplished by the 3D network formed by HMP and LMP through hydrogen bonding and calcium-mediated ionic cross-linkages, respectively. Further, the pectin gel continues to swell in the neutral conditions (pH 7.4) of intestinal and colonic fluids without disintegrating the gel matrix. Günter et al. (2019) showed that the orally administered pectin gel microparticles underwent gradual swelling in the simulated gastric and intestinal fluids. The resultant reduction in food intake during the first five hours of free-feeding was attributed to the increased water-binding and swelling capacity of stomach digesta (Tan et al. 2017). Further, the pectin micro-particles can swell without losing the gel structure under simulated intestinal conditions.

Pectin's ability to induce satiety by delaying gastric emptying has been tested on mice (Khramova et al. 2019). Low-methoxyl apple pectin with (AP-Ca) and without (AP) the supplementation of calcium ions was orally administered to the test mice. Control mice were fed with water, for comparison. During the first 30 min after feeding, the weight of gastric digesta was reduced by 50% in the control mice fed with water. Contrastingly, in mice orally administered with AP and AP-Ca, the weight of gastric digesta decreased by 75% and 50%, respectively, after 60 min and 90 min of feeding. Thus, AP and AP-Ca orally administered to mice before feeding delayed the gastric emptying by one-half time.

The mere delaying of gastric emptying caused by pectin gel does not ensure satiety induction. For instance, pectin gel beads derived from commercial pectin hydrated and swelled in acidic gastric conditions without significant damage to the gel matrix. But, in simulated intestinal fluid, the intensively swollen pectin gel beads eroded and eventually dissolved within the first few hours of incubation, attributed to the higher degree of esterification, lower initial calcium content and the presence of sodium, potassium and phosphate ions in the intestinal fluids. The presence of ions may cause the release of calcium ions from the pectin chains.

**Table 2.** Applications of pectin gel in the encapsulation of food bioactives.

S. No.	Source of pectin	Type of pectin	Core compound	Processing condition		Major findings	References
				Method of gelation	Formulation		
1.	Citrus peel	<ul style="list-style-type: none"> <li>High methoxyl pectin</li> <li>DE: &gt;74%</li> </ul>	Iron	Ionic gelation	<ul style="list-style-type: none"> <li>Pectin solution in acetic acid-sodium acetate: 4% vol/wt</li> <li>FeSO<sub>4</sub> solution: 150 mM</li> <li>1:1 alginate pectin solution with 7.81% (wt/wt) anthocyanin</li> <li>Buffers 0.1 M pH 1.2 HCl/KCl or 0.1 M pH 3.0 citric acid/sodium citrate</li> </ul>	<ul style="list-style-type: none"> <li>Stable beads were formed with essential trace mineral and dietary fiber</li> <li>Remedy for iron deficiency</li> <li>Blueberry was encapsulated at a higher efficiency than purple corn</li> <li>Increased concentration of alginate and gum improved the encapsulation efficiency</li> <li>Encapsulation in alginate-pectin gel reduced 90% loss of anthocyanins under dark conditions</li> <li>Pectin hydrogel increased the tolerance of <i>L. plantarum</i> in the acidic media</li> </ul>	Ghibaudo et al. (2018)
2.	-NM-	<ul style="list-style-type: none"> <li>High methoxyl pectin</li> <li>DE: 71%–75% and 63%–67%</li> </ul>	Anthocyanins from purple corn and blueberry extracts	Stock solution was extruded through a needle with inner diameter of 0.337 mm, using a peristaltic pump to a gently agitated buffer solution.			Guo, Giusti, and Kaletunc (2018)
3.	-NM-	<ul style="list-style-type: none"> <li>Low methoxyl pectin</li> <li>DE: 28%</li> </ul>	<i>L. plantarum</i> cells	Extrusion method (external gelation method)	<ul style="list-style-type: none"> <li>Pectin solution: 0.5%–2%</li> <li>Starch solution: 0%–1.5%</li> <li>CaCl<sub>2</sub>: 0.2 M</li> <li><i>L. plantarum</i> cells: 10.01 log CFU/g</li> </ul>		Dafe et al. (2017)
4.	-NM-	<ul style="list-style-type: none"> <li>Low methoxyl pectin</li> <li>DE: 50%</li> </ul>	$\alpha$ -tocopherol	Ionotropic gelation	<ul style="list-style-type: none"> <li>Pectin solution: 2% wt/vol</li> <li>Glycerin: 1% vol/vol</li> <li>Tween-80 (emulsifier): 0.1% wt/vol</li> <li><math>\alpha</math>-tocopherol: 1% wt/vol</li> <li>CaCl<sub>2</sub> solution: 5% wt/vol</li> </ul>	<ul style="list-style-type: none"> <li>Microencapsulation of <math>\alpha</math>-tocopherol was successfully accomplished with pectin-alginate gel</li> <li>Encapsulated <math>\alpha</math>-tocopherol can be used as antioxidants in fat-based products</li> <li>Encapsulation increased the stability and ease of handling of the product</li> <li>The beads with garlic and holy basil essential oils showed good antimicrobial properties</li> </ul>	Singh, Kaur, and Kumar (2018)
5.	Citrus peel	<ul style="list-style-type: none"> <li>Low methoxyl pectin</li> <li>DE: 2.9%</li> </ul>	Garlic and holy basil essential oils	Ionotropic gelation	<ul style="list-style-type: none"> <li>Chitosan: 0.2%–0.7% wt/vol</li> <li>Pectin: 3.5%–5.5% wt/vol</li> <li>CaCl<sub>2</sub>: 5%–20% wt/vol</li> </ul>	<ul style="list-style-type: none"> <li>The optimal bead composition was 0.3%–0.6% wt/vol chitosan, 3.9%–5.1% pectin, and 8.0%–17% CaCl<sub>2</sub> and exhibited a very good encapsulation efficacy (62.16%–79.06%) and cumulative release efficiency (31.55%–37.81%)</li> <li>The antimicrobial property increased with increase in bead weight</li> </ul>	Torpol et al. (2019)
6.	Citrus peel	<ul style="list-style-type: none"> <li>Methoxyl groups: <math>\geq</math>6.7</li> </ul>	<i>Lactococcus lactis</i>	Ionotropic gelation	<ul style="list-style-type: none"> <li>Alginate-to-pectin ratio: 100:0; 75:25; 50:50; 25:75; 0:100</li> <li>CaCl<sub>2</sub>: 100 mM</li> </ul>	<ul style="list-style-type: none"> <li>A 75:25 alginate-pectin ratio enriched with glucose resulted in hydrogel beads with best mechanical properties than pure alginate and pectin and provided desirable protection to <i>L. lactis</i></li> <li>FTIR spectroscopy confirmed possible interactions between alginate and pectin during inter-penetrating network formation</li> <li>Resulted in best release of nisin during the storage period</li> <li>Based on the cell viability and proliferation, optimal formulation for the preparation of calcium pectinate beads was found to be pectin of DE 35%, pectin concentration: 2.3% and calcium concentration: 100 mM</li> </ul>	Bekhit et al. (2016)
7.	-NM-	<ul style="list-style-type: none"> <li>Low-methoxyl pectin</li> </ul>	C3A cells	Ionotropic gelation	<ul style="list-style-type: none"> <li>Pectin solutions at concentration of 0.5%–5% were prepared in 0.9% NaCl solution</li> <li>CaCl<sub>2</sub>: 100 mM</li> <li>C3A cells: 10<sup>6</sup> cells/ml pectin solution</li> </ul>	<ul style="list-style-type: none"> <li>Interaction effects between pectin and calcium and the linear contribution of pectin concentration influenced the breakage rates of calcium pectinate beads</li> </ul>	Zhao, Zhang, et al. (2018)

(continued)

**Table 2.** Continued.

S. No.	Source of pectin	Type of pectin	Core compound	Processing condition			Major findings	References
				Method of gelation	Formulation			
8.	Citrus fruits	• Low-methoxyl pectin	<i>Lactobacillus plantarum</i> and DHA fatty acid (coencapsulation)	Ionotropic gelation	<ul style="list-style-type: none"> <li>Pectin: 0%–1.3%</li> <li>Alginate: 0.7%–2%</li> <li>Fish gelatin: 0%–1.3%</li> <li>CaCl<sub>2</sub>: 5% wt/vol</li> <li>DHA rich-oil: 1.5% wt/vol</li> <li>Cell suspension</li> </ul>	<ul style="list-style-type: none"> <li>The optimal formulation was found to be 1.06% alginate, 0.55% pectin and 0.39% gelatin</li> <li>Survivability of microencapsulated cells was found to be 88.66%, compared to that of free cells (50.36%)</li> <li>DHA increased smoothness and compactness of the surface of particles</li> <li>Thermal stability of DHA-loaded capsules was high compared to unloaded ones</li> </ul>	Vaziri et al. (2018)	

-NM:- details not mentioned.

Since calcium ions are the cross-linking points of pectin, the release of the same can result in increased water uptake by the pectin gel microparticles (Remuñán-López and Bodmeier 1997). Several approaches have been employed to increase the stability of pectin gel under gastrointestinal conditions. These include coating the pectin gel with materials such as hydroxypropylmethyl cellulose and tailoring the structure of pectin granules at the isolation stage to obtain different swelling and biodegradability properties under gastrointestinal conditions (Mundargi, Rangaswamy, and Aminabhavi 2011; Popov et al. 2017).

The delayed gastric emptying caused by the pectin gel under the acidic conditions of the mouse stomach was governed by the rheological properties of gastric digesta formed in the postprandial period (Khramova et al. 2019). Gastric digesta of mice was identified as shear-thinning or pseudoplastic type of fluid (viscosity decreases with an increase in shear rate). After the oral administration of water, AP, and AP-Ca, the viscosity of gastric digesta was monitored during the interval from 5 to 90 min. During gastric digestion, the viscosity of the gastric digesta of mice orally administered with water decreased by 78%. On the contrary, AP-Ca gelled in the stomach's acidic conditions and increased the viscosity of the mice gastric digesta, right from 5 min after feeding. The viscosity of gastric digesta increased with AP as well, but only after 30 min. Early-onset of an increase in the viscosity of gastric digesta of mice fed with AP-Ca was attributed to the supplementation of pectin solution with CaCO<sub>3</sub>.

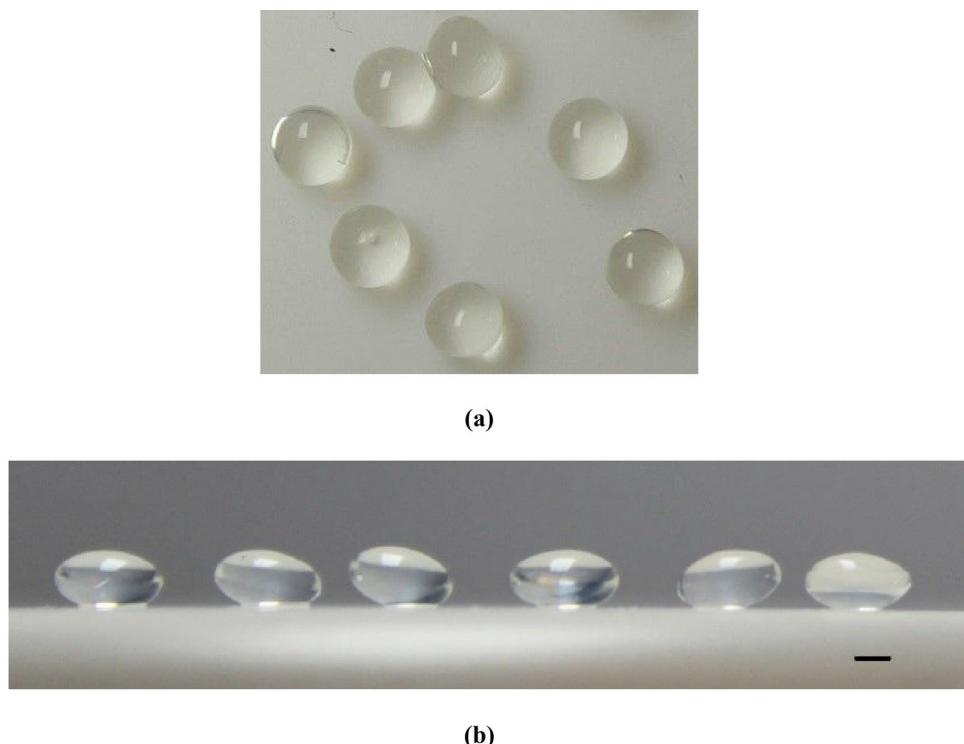
AP-Ca formed a gel bolus in the stomach by developing ionic cross-linkages via calcium (Morris et al. 1982) and provided a more significant increase in the viscosity values of mice gastric digesta compared to AP. Thus, at any point of time, the viscosity of gastric digesta of mice orally administered with water, AP, and AP-Ca was found to increase in the order of water < AP < AP-Ca (Khramova et al. 2019). A similar trend was observed with the elastic or storage modulus ( $G'$ ) of the gastric digesta, which was at ~280–3100 Pa for the gastric digesta of mice orally administered with AP-Ca and at ~150–950 and ~100–800 Pa for gastric digesta of mice fed with AP and water, respectively.

Further, the increase in viscosity of the gastric digesta caused by the oral administration of AP and AP-Ca seemed

to slow down the digesta mixing, the interaction between digestive enzymes and nutrients, and the breakdown of complex nutrients into absorbable components. Consequently, the absorption of glucose and other nutrients was slowed down (Chutkan et al. 2012). Prevention of rapid glucose absorption by pectin gel is facilitated in the intestine, wherein it holds glucose molecules inside the chyme. Also, pectin gel retained in the intestine restricts the interaction between luminal contents and digestive enzymes and thickens the unstirred water layer to decrease the diffusion and uptake of glucose (Kaczmarczyk, Miller, and Freund 2012). Thus, gelling of pectin in the intestine is considered central to the pectin-mediated reduction in weight gain and fatty liver (by 50%) (Bray et al. 2018).

Another mechanism by which pectin gels reduce the postprandial blood glucose concentration is the negative feedback loop provoked by hormonal regulation. These regulatory mechanisms are complex as they involve the central nervous system, the vagus nerve, and neurohumoral peptides (Hansen 2003). Several neurohumoral gut peptides are secreted from enteroendocrine cells in the gastric and intestinal mucosa during the fasting and postprandial periods. The examples include ghrelin, cholecystokinin (CCK), glucagon-like peptide-1 and 2 (GLP-1, GLP-2) and peptide YY (PYY). Among the above peptides, CCK, PYY, and GLP-1 are responsible for delaying the gastric transit rate via central signaling. These peptides prolong gastric emptying by modulating the gastric and intestinal motility by activating the receptors on sensory, vagal, and intrinsic afferent neurons (Steinert et al. 2017).

Pelkman et al. (2007) developed a low-energy beverage from calcium-gelled alginate-pectin to control food intake in overweight and obese women. Each dose of the beverage was designated to provide 40 kcal. On consumption, the alginate-pectin beverage delayed the absorption of nutrients by gel lumps and thereby increased the gastric volume and delayed post-consumptive effects. The beverage's satiation effect was proposed to be promoted by the stimulation of incretin responses caused by the delayed absorption. This study recommended that consuming calcium-gelled alginate-pectin twice per day can reduce spontaneous food intake in



**Figure 8.** Smart pectin hydrogels: (a) spherical-shaped hydrogel particles and (b) disc-shaped hydrogel particles (Reproduced with permission from and Kaletunç, 2016).

obese women. Foods containing calcium-gelled alginate that is activated in the stomach may be useful for weight loss.

### Recent developments in pectin hydrogel research

In recent years, pectin hydrogel research has shifted toward tailoring the dimensions and microstructure to achieve certain innovative features and applications. These include the development of so-called “*smart gels*” (Ullah et al. 2015), “*injectable hydrogels*” (Moreira et al. 2014), “*aerogels*” (Zhao, Malfait, et al. 2018), “*xerogels*” (Singha et al. 2017), and “*oleogels*” (Luo et al. 2019). A few case studies on these recent trends in pectin hydrogel research are discussed in the subsequent sections.

### Smart pectin hydrogels

Smart hydrogels exhibit a significant physiochemical change in response to even a slight variation in their surroundings. But, these changes are reversible, and hence the hydrogels can return to their initial state once the stimulus is removed after a reaction. The stimulus may be physical (ex. light, pressure, temperature) or chemical (ex. pH, enzymes, ionic factors, and chemical agents (Bacelar et al. 2017; Mantha et al. 2019). In food systems, pH-responsive smart hydrogels have been used as vehicles for the targeted delivery of bioactive compounds. For instance, composite hydrogel formed from a mixture of alginate (A) and pectin (P) was extruded under low pH conditions to generate spherical (Figure 8a) and disc-shaped (Figure 8b) particles. Different shapes were obtained by varying the distance between the needle and surface of the buffer bath during particle production. Purple

corn (PC) and blueberry (BB) extracts were encapsulated in these hydrogel particles to protect the bioactive anthocyanins (Guo and Kaletunç 2016).

Under simulated conditions of the gastrointestinal tract, the hydrogel particles were stable at the stomach’s acidic pH (3.0). They were found to release the anthocyanins when triggered by an increase in pH at the target site (lower GI tract). At pH 5.0 and pH 7.0, hydrogel particles underwent a gel-sol transition and dissolution due to increased electronic repulsive forces leading to the release of bioactive compounds from the matrix. The authors proposed the applications of these pH-responsive hydrogel particles in low-pH beverages (pH below 3.0). This was proved by monitoring the diffusion or leakage of anthocyanins from the hydrogel particles to the surrounding solution, a pH 3.0 buffer to mimic a low pH beverage. The stability over 25 days was found to improve at low temperature and high particle weight-to-solution volume ratio. During storage, spherical hydrogel particles retained a higher amount of anthocyanins (36.6%) than the disc-shaped particles (6.5%), due to the lower surface area of the former ( $22.6 \text{ mm}^2$ ) than the latter ( $25.7 \text{ mm}^2$ ), at a constant volume (Guo, Giusti, and Kaletunç 2018).

Similarly, a pectin-gelatin hydrogel layer containing microparticles of antimicrobial agents such as pectin-nisin and potassium sorbate was used as a smart and active packaging system to control weak syneresis, microbial spoilage, and oxidation in addition to intelligent detection in case of severe syneresis. The microparticles of antimicrobial agents were enclosed in the discontinuous phase of the pectin-gelatin network to facilitate controlled-release. This smart system leveraged the limited ability of the pectin-gelatin composite to imbibe water. As a result, when over-swelling

occurred, the pectin-gelatin gel disintegrated and dissolved. This change in state or volume could be readily visualized or with color or turbidity indicators. Thus, the active pectin-gelatin hydrogel layer transformed into a smart absorber layer is capable of indicating the severe quality loss. Further, this product could control microbial spoilage to some extent, as it is incorporated with antimicrobial agents (Haghghi and Mousavi 2012).

### **Injectable pectin hydrogels**

Injectability is a behavior that confers protection to the bioactive molecules or cells against mechanical damage during extrusion through a needle. Generally, injectable hydrogels are prepared by injecting small aliquots of a nutraceutical-biopolymer solution into a gelling solution that contains specific components that promote cross-linking of the biopolymer chains (ex. mineral ions, acids, bases, or enzymes) (McClements 2017). Being an anionic polysaccharide, pectin demonstrates innate biocompatibility and soft tissue-like behavior, which renders it suitable for the production of injectable systems. The presence of calcium-induced cross-linking modulates the viscoelastic properties and thereby the injectability of gels. The slow cross-linking kinetics of injectable pectin hydrogels favors *in situ* gel-formation for injectable therapeutic delivery of drugs, proteins, or cells (Gutowska, Jeong, and Jasionowski 2001). The acidic pH of pectin gels can restrain their usefulness as injectable systems. Their pH can be increased by using different bases such as NaOH, NaHCO<sub>3</sub>, and TEA (tris-ethanol-amine). NaHCO<sub>3</sub> is preferred as it raises the pH of gels without causing depolymerization of pectin.

Usually, pectin-based injectable systems are tailored to possess a shear-thinning behavior, i.e., low viscosity at high shear to enable the injection through a needle (Moreira et al. 2014). Indeed, the injectability of a hydrogel is confirmed by rheological analyses. At physiological temperatures (i.e., 35–42 °C), elastic modulus and complex viscosity of the hydrogels remained stable, thus confirming their injectability. The intended application of these injectable hydrogels is to provide a microenvironment for the delivery of bioactive molecules. But, hitherto, injectable pectin hydrogels have been predominantly used in tissue engineering and drug delivery, and their food applications are still at the nascent stage. Hence, the potential scope of research exists in this area to prepare injectable pectin hydrogels of different sizes, structures and functional properties to encapsulate bioactive components.

### **Aerogels**

Aerogel is any material that is less-dense, sol-gel derived, and predominantly mesoporous, with its pore diameter ( $\varnothing$ ) ranging between 2 and 50 nm (Singha et al. 2017). However, this definition is different from that specified by IUPAC, which requires aerogels to be strictly microporous ( $\varnothing < 2$  nm) (Aegerter, Leventis, and Koebel 2011; IUPAC 2014). Due to its gelling and film-forming abilities, pectin is considered an ideal candidate for the preparation of freeze-



**Figure 9.** Photograph of pectin/clay aerogel composite (Reproduced with permission from Chen et al. 2013).

drying-based aerogels with potential applications in foods. The major application of pectin aerogel has been realized in food packaging, owing to its proven biodegradability.

To achieve cross-linking during the preparation of pectin aerogel, monovalent ( $\text{Na}^+$ ), divalent ( $\text{Ca}^{2+}$ ), and trivalent ( $\text{Al}^{3+}$ ) cations were added to the aqueous pectin solution. Apart from cross-linking, the addition of cations promoted the supramolecular bonding of chains and improved their mechanical properties. The cross-linking ability of cations was in the order of  $\text{Ca}^{2+} > \text{Al}^{3+} > \text{Na}^+$ . Usually, it is expected that the use of trivalent cations would lead to a higher extent of pectin cross-linking than that attained with the divalent  $\text{Ca}^{2+}$  ions (Chen et al. 2013). However, it was not the case observed in this study. This may be because cross-linking is a diffusion-controlled process, which is a function of the ionic size.  $\text{Al}^{3+}$  ions have been found to have a smaller ionic size compared to calcium, due to which they might have merely diffused through the aerogel without cross-linking (Reddy 2016). One more reason for the reduced thermal stability of aluminum-cross-linked pectin could be the bonding water in the aluminum sulfate additive, which led to a continuous weight loss during heating from 86.5 °C to 250 °C. In addition to cations, sodium montmorillonite ( $\text{Na}^+$ -MMT) was also added to the aerogel forming pectin solution to improve its mechanical properties. Increasing the clay content in pectin aerogels increased the overall bulk densities, and the corresponding improvement in mechanical properties was significant. For instance, when 2.5% (wt/wt) clay was added to 5% (wt/wt) pectin solutions, the modulus of aerogels increased from 10 to 330 kPa. Finally, pectin aerogel of diameter 2 cm (Figure 9) was obtained using the freeze-drying process (Chen et al. 2013).

The biodegradability of pectin aerogels and pectin/clay aerogels was studied in compost media at room temperature. Wheat starch was chosen as a reference due to its effective biodegradation (~60% after 30 days). Pectin aerogels and pectin/clay aerogels were found to possess higher biodegradability than wheat starch. While wheat starch reached only 30% biodegradation after 30 days, pectin aerogels and pectin/clay aerogels attained 40%–57% after ten days of immersion in the compost media. The presence of clay and multivalent cations enhanced the hydrophilicity of pectin aerogels to improve their biodegradation rates. As water is an essential requirement for microbial growth, samples with high moisture aided

the propagation of microorganisms, which in turn increased the biodegradation rate. It was also hypothesized that the presence of clay and salt in the pectin aerogel might alter the integrity of pectin aerogel to cause more defects and thereby enhance the biodegradation process (Chen et al. 2013).

Pectin-based nanocomposite aerogels were prepared using tert-butanol and zinc ions as cross-linking agents. The cross-linking reaction took place through slow diffusion of the cross-linking solution through the pectin dispersions. After complete gelation, the gels were dried using supercritical CO<sub>2</sub> to obtain the aerogel. Further, the addition of TiO<sub>2</sub> nanoparticles increased the strength and rigidity (tensile strength and Young's moduli) of pectin aerogels due to the stronger intermolecular forces and denser chain packaging. The tensile strength of pectin gel-based nanocomposites that had 20% (wt/wt) of TiO<sub>2</sub> was 6-fold higher than the control pectin aerogel (Nešić et al. 2018). Similarly, Young's moduli of the composite pectin-TiO<sub>2</sub> aerogels (9.8–33 MPa) were also higher than that of the control pectin aerogel (7.5 MPa). The higher interfacial interaction between pectin chains and TiO<sub>2</sub> nanoparticles established through the formation of hydrogen bonds led to improved mechanical properties.

Notably, pectin aerogel without TiO<sub>2</sub> exhibited the lowest value of the thermal conductivity for (0.022 W m<sup>-1</sup> K<sup>-1</sup>), which was even lower than that of cellulose and polystyrene foams (0.046–0.054 and 0.030–0.040 W m<sup>-1</sup> K<sup>-1</sup>, respectively) that are presently used as flexible thermal insulators commercial food packaging. However, the TiO<sub>2</sub> containing aerogels showed a slight increase in thermal conductivity (0.025 at 20% concentration of TiO<sub>2</sub>). Thus, pectin-based aerogels have potential applications as super-insulators for the packaging of temperature-sensitive foods. This is relevant as the packaging material should have exceptional insulating properties to maintain the quality and improve the shelf-life of perishable foods (Nešić et al. 2018).

### Xerogel

Like aerogel, "xerogel" is a dry gel prepared by evaporating the liquid in the pores of the hydrogel at ambient temperature. Nevertheless, the xerogels vary from aerogels in terms of their pore size and density (Scherer 2001). The rationale of drying the pectin hydrogel beads is to improve their stability in solution and facilitate easy handling and storage of the beads. Pectin-based xerogel beads have been found to possess good stability, irrespective of the pH and stirring conditions (Mata et al. 2010). Pectin xerogels prepared from sugar-beet pulp have been utilized for the biosorption and removal of heavy metals such as copper, lead and cadmium from aqueous solutions. The use of pectin xerogels for biosorption is relevant as heavy metals are potentially unmanageable pollutants of water resources, the concentration of which can increase with time in living tissues all along the food chain.

Molecular scale Fourier transform infrared (FTIR) spectroscopy was employed to identify the possible functional groups in HMP and LMP that can bind heavy metals in an aqueous solution. The results revealed that the excessive and free anionic carboxylic acid groups in HMP and LMP are

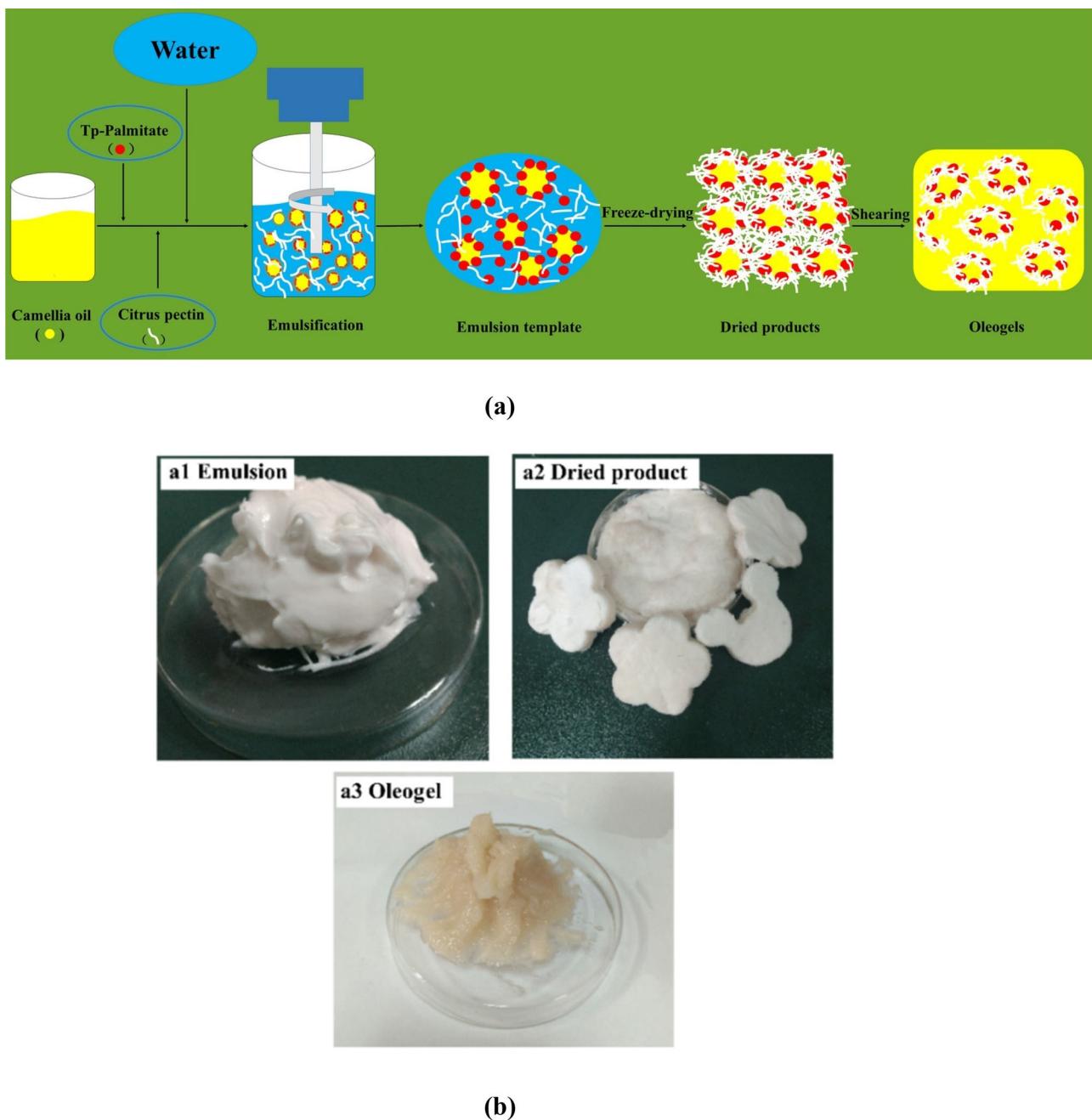
responsible for absorbing heavy metal cations (Mata et al. 2010; Jakóbik-Kolon et al. 2017; Khotimchenko, Kovalev, and Khotimchenko 2007). The major mechanism of biosorption was identified as ion exchange between calcium in the pectin xerogel structure and chelation or complexation with carboxyl groups. During the biosorption, calcium in the gel is replaced by the cations of cadmium, lead and copper, which causes a restructuring of the gel matrix. The pectin xerogels were found to have a compact structure and higher stability than the pectin hydrogels (Mata et al. 2009a). Further, the pectin xerogels exhibited superior biosorption/uptake kinetics for heavy metals compared to alginate xerogels under similar experimental conditions (Mata et al. 2009b).

### Oleogels

"Oleogelation" is an oil structuring approach to transform a liquid-oil into a "gel-like" viscoelastic structure having liquid-oil at a concentration of more than 90% (wt/wt) (Abdallah, Sirchio, and Weiss 2000; Botega et al. 2013; Patel et al. 2013). It is a recent technique for developing saturated fat-free food products and healthy solid-fat substitutes (Marangoni and Garti 2018). A critical factor that determines the practicality of this process is the selection of structurants for oil gelation (Botega et al. 2013). In this context, recent studies have identified the potential of high methoxyl pectin (HMP) as a candidate structurant for oleogelation due to its ability to adsorb at the oil-water interface while forming a gel in the water phase (Chan et al. 2017; Schmidt, Schütz, and Schuchmann 2017; Verkempinck et al. 2018).

High-methoxyl citrus pectin in combination with tea polyphenol (Tp)-palmitate particles were used to convert liquid camellia oil into structured edible oleogels using the "emulsion-templating" approach (Figure 10a). The water-in-oil emulsion of Tp-palmitate in camellia oil (2.5% wt/vol) was flash-cooled, followed by the addition of citrus pectin (1.5%–4.5% wt/vol). Then, the emulsion-templates (Figure 10b [a1]) were prepared by adding the above dispersion (60 mL) into distilled water (40 mL) and emulsifying the same using a high-speed shear emulsifying machine at 8 ± 2 °C and 20000 RPM for 2 min. Further, the emulsion-templates were dried in a freeze-dryer to remove the water phase (Figure 10b [a2]). Eventually, the camellia oil oleogel was obtained by shearing the freeze-dried products using an electric mixer at 10000 rpm for 2 min (Figure 10b [a3]) (Luo et al. 2019).

The concentration of citrus pectin significantly influenced the physical properties of emulsions, dried products, and oleogels. This was attributed to the reinforcement from the interactions and entanglements of the polymer. As the pectin concentration increased, the following changes were observed: (1) increase in the stability and viscoelasticity of emulsions; (2) increase in the hardness of dried products having a more compact structure and (3) increase in the oil binding capacity and gel strength of the oleogels. Pectin oleogels revealed a good gel strength, evident from their high value for elastic modulus ( $G' > 17,000$  Pa) and a good thixotropic recovery at pectin concentration higher than 1.5% (wt/vol). Moreover, the tea polyphenols contributed to

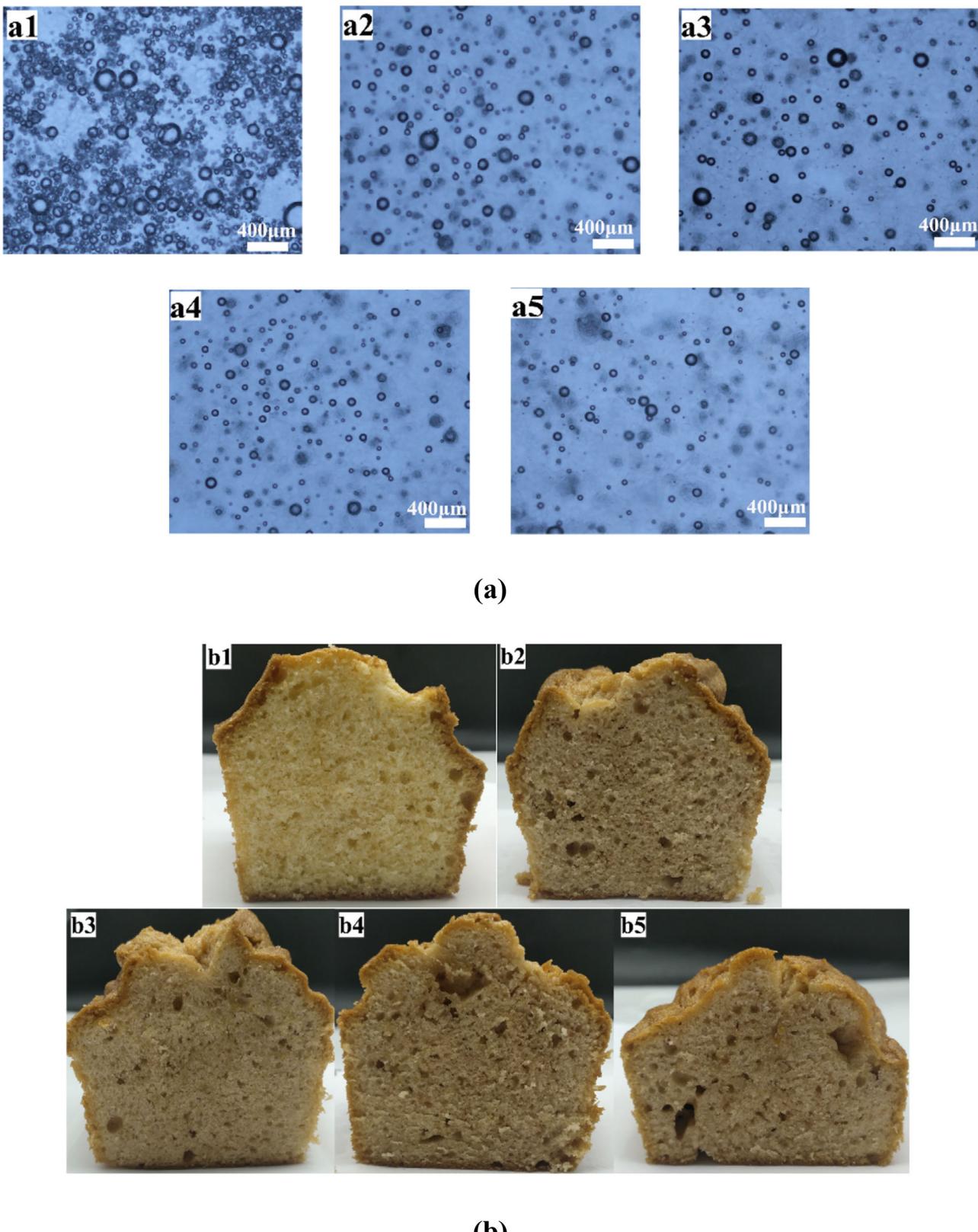


**Figure 10.** Pectin oleogel: (a) schematic of the emulsion-templating approach; (b) visual appearance of (a1) emulsion; (a2) dried product and (a3) oleogel (Reproduced with permission from Luo et al. 2019).

the high antioxidant activity of oleogels. This is in accordance with the similar activity exhibited by pectin composite films incorporated with tea polyphenols, as explained earlier.

Further, oleogels were incorporated in cakes as a replacement for butter. The bubble count in cake batter is central to the final product's quality (Patel, Cludts, et al. 2014). The number of bubbles in the oleogel containing batter was less than that in the batter with butter (Figure 11a). The absence of fat crystals and emulsifiers in the oleogels was suggested as the reason for the reduced bubble count (Patel, Cludts, et al. 2014; Patel, Rajarethinem, et al. 2014). The bubble count also showed a significant decrease with the increase in the concentration of the citrus pectin in oleogels. The authors proposed that the batter containing oleogels with a

higher concentration of pectin would have greater mechanical strength, which might have constrained the air occlusion into the batter during mixing. The product with oleogels was subjected to sensory evaluation in comparison with that of cakes with butter. The oleogel containing cakes obtained a hedonic score of 21.49–27.58 for the overall quality of cakes compared to 32.03 for cakes with butter. The oleogel-containing cakes were slightly harder than the cakes prepared with butter (Figure 11b). Nevertheless, this study opened up a new possibility for the partial replacement of butter with camellia oil-based oleogels. This approach can potentially reduce the trans and saturated fatty acids to improve the nutritional profile of high-fat products such as baked foods (Luo et al. 2019).



**Figure 11.** Light microscopy images of cake batters and photographs of cakes: (a) light microscopy images of cake batters prepared using butter (a1), oleogels with 1.5% (a2), 2.5% (a3), 3.5% (a4) and 4.5% (a5) (m/v) citrus pectin, respectively; (b) photographs of cake products prepared using butter (b1), oleogels with 1.5% (b2), 2.5% (b3), 3.5% (b4) and 4.5% (b5) (m/v) citrus pectin, respectively (Reproduced with permission from Luo et al. 2019).

Besides the above, the scope for future research in this discipline would be to develop pectin xerogels for the bio-sorption of processing-induced toxic compounds. Pectin-based fluorescent or light-emitting gel is also an area with

potential scope for investigation. Fluorescent polymer gels that change in color on heating, exposure to the acidic environment or on disruption can potentially function as sensors to detect changes in processed foods resultant from

time-temperature abuse during storage. With these expected advancements, pectin hydrogel is likely to be an essential food ingredient that would be widely used for various functions in the days to come.

## Conclusions

In recent years, food consumers prefer clean label products with nutritional wellness, convenience, and variety, but without compromising taste and quality. Consequently, food manufacturers require natural ingredients with novel applications capable of resulting in superior-quality products. Based on the insights presented in this review, it is obvious that pectin hydrogel is a fascinating food ingredient or a product by itself, owing to the intricate relationship between its dimensions, structure, composition and rheology. With the current level of advanced knowledge on pectin production and gelation, the advent of pectin hydrogels with new utilities in the food, nutraceutical and allied fields is obvious. In this context, pectin hydrogel research is certainly envisaged to go through many advancements in the future. Further research on engineered and functional pectin hydrogels would be a significant addition to the current state of the art on soft food materials and their role in achieving sustainable food systems.

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