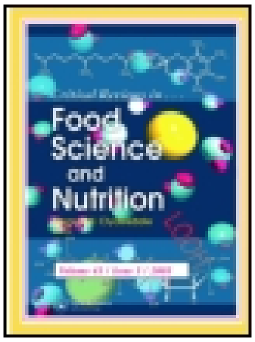

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REVIEW

3D food printing: Applications of plant-based materials in extrusion-based food printing

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ABSTRACT

As an emerging digital production technology, 3D food printing intends to meet the demand for customized food design, personalized nutrition, simplification of the food supply chain system, and greater food material diversity. Most 3D food printing studies focus on the development of materials for extrusion-based food printing. Plant-based foods are essential for a healthy diet, and they are growing in popularity as their positive effects on human health gain wider recognition. The number of original studies on plant-based printable materials has increased significantly in the past few years. Currently, there is an absence of a comprehensive systematic review on the applications of plant-based materials in extrusion-based food printing. Thus, this review aims to provide a more intuitive overview and guidance for future research on 3D printing of plant-based materials. The requirements, classifications, and binding mechanisms of extrusion-based food printing materials are first summarized. Additionally, notable recent achievements and emerging trends involving the use of plant-based materials in extrusion-based food printing are reviewed across three categories, namely, hot-melt (e.g., chocolate), hydrogel, and soft (e.g., cereal- and fruit/vegetable-based) materials. Finally, the challenges facing 3D food printing technology as well as its future prospects are discussed.

KEYWORDS

Additive manufacturing; chocolate; extrusion-based food printing; fruit- and vegetable-based materials; hydrogels; cereal-based foods

Introduction

Three-dimensional (3D) printing, also known as additive manufacturing, uses computer-aided design software to control the layer-by-layer construction of 3D objects (Jiang et al. 2019). 3D printing technology has several advantages, such as freedom of design, mass customization, minimal waste, complex structure fabrication, and rapid prototyping, which have made it attract increasing interest recently (Ngo et al. 2018). As an emerging digital technology, 3D printing has been adopted in diverse industries, including mechanical engineering, aerospace, biomedical engineering, pharmaceutical industry, biological engineering, and food processing (Ngo et al. 2018). In particular, the last decade has witnessed a considerable increase in the application of 3D printing within the food industry.

3D printing was first introduced into the food sector using extrusion-based printers (Fab@home) by researchers at Cornell University (Periard et al. 2007). The potential of 3D food printing in customizing food design, personalizing nutrition, simplifying the food supply chain, and expanding food material sources has attracted a devoted research base (Liu et al. 2017). At present, four types of 3D printing methods can be applied to food printing: selective laser sintering, extrusion-based printing, binder injection, and inkjet printing (Sun et al. 2015), among which extrusion-based food printing is the most established. Recent research has led to the development of many extrusion-based food printing materials, including chocolate (Mantihal et al. 2017), dough (Yang, Zhang, Prakash, et al. 2018), mashed potatoes (Liu, Zhang, et al. 2018), powdered sugar (Holland et al. 2018), cheese (Le Tohic et al. 2018), surimi gel (Wang et al. 2018), insect-enriched snacks (Severini, Azzollini, et al. 2018), and fruits and vegetables (Severini, Derossi, et al. 2018; Yang, Zhang, Bhandari, et al. 2018).

The quality of printed food is determined by material properties, processing factors, and post-processing, with most studies focused on refining these aspects. As successful printing is contingent on the material properties, these are the most important factors affecting the printing precision (Feng, Zhang, and Bhandari 2019). Therefore, the development of novel printable food materials has become a hot research topic. Indeed, the number of original studies has proliferated remarkably in the last three years, greatly

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expanding the scope of printable food materials. Relatively few of these studies have reported on animal-based materials, with the majority focusing on plant-based materials. Therefore, this article provides a detailed and timely discussion on the advances made in printable plant-based food materials research, which clarifies notable achievements in this field and outlines the directions of future research.

This paper begins with an overview of extrusion-based edible printing materials before reviewing the research status of plant-based materials in extrusion-based food printing, including chocolate, hydrogels, cereal-based foods, and fruit- and vegetable-based materials. Finally, the most notable challenges and trends shaping the future of research on 3D food printing technology are discussed. Additionally, this review proposes ideas for the application of plant-based materials in 3D food printing technology, the rational design of printable materials, and the acceleration of plant-based printing material developments.

Requirements, classification, and binding mechanisms of extrusion-based food printing materials

Material requirements

Owing to the complex composition of food materials and the differences in physical and chemical properties of each component, only select materials are suitable for 3D food printing, which can make its application challenging. Materials used for 3D food printing must meet three essential requirements: printability, applicability, and suitability for post-processing (Godoi, Prakash, and Bhandari 2016; Nachal et al. 2019).

Printability refers to the ease with which the material can be controlled and deposited by a 3D printer, including shape retention after deposition. Materials with good printability can be used to fabricate complex structures. Printing materials should have specific physicochemical, rheological, and mechanical properties, with different printing technologies requiring different material properties. For example, in extrusion-based food printing, the moisture content, rheological properties, specific crosslinking mechanisms, and thermal properties (melting point and glass transition temperature) play critical roles (Liu et al. 2017).

Applicability refers to the ability of the material to fulfill certain functions. Owing to its ability to build complex geometric designs and structures, 3D printing is attractive for many applications. In addition to the malleability associated with printability, printing materials must satisfy further food-related needs. For example, the specific nutritional requirements of a given population can be met by incorporating nutritional value into unique structural designs. The feasibility of any application depends on the properties of the supplied materials and the scalability of production (Nachal et al. 2019). Therefore, the applicability of food materials determines the scope of 3D printing-based technology in the food sector.

Post-processing behavior refers to the ability of a material to undergo further processing after printing. Although some foods can be eaten directly, most printed foods undergo some form of cooking prior to consumption (Nachal et al. 2019), making post-processing quality vital for consumer acceptance. Researchers compared two post-processing drying methods (oven drying and freeze drying) and found that printed samples with an initial dry matter content of $\leq$ 35% experienced significant shrinkage during oven drying. In contrast, for samples with an initial dry matter content of $\geq$ 45%, the sample shape was preserved after each drying method (Lille et al. 2018). This is mainly because a lower water content corresponds to a higher initial structural strength. It is crucial to select materials with appropriate rheological and mechanical properties to endure the cooking process without incurring damage.

Material classification

In general, food materials can be divided into three categories according to their printability: natively printable materials, non-printable traditional food materials, and alternative ingredients (Dankar, Haddarah, et al. 2018; Dick, Bhandari, and Prakash 2019). Natively printable materials are extruded easily from the printing nozzle without the addition of additives (examples include hydrogels, cake frostings, soft cheese, hummus, and chocolate). Such materials exhibit high stability and can maintain the printing shape after deposition without (normally) requiring post-processing (Dankar, Haddarah, et al. 2018). Alternatively, traditional foods such as rice, meat, fruits, and vegetables are essentially non-printable materials, and they require the use of additives to improve their rheological properties and make them printable. For example, Cohen, Lipton, and Cutler (2009) added xanthan gum and gelatin to improve the material properties of natural foods (vegetables, fruits, and meat), thereby obtaining a more desirable printing structure capable of maintaining a satisfactory shape after deposition. Furthermore, materials rich in protein, dietary fiber, and bioactive substances, such as algae, fungi, lupine, and insects, are often used as alternative ingredients in food printing. For instance, in the “Insect Au Gratin” project, insect powders mixed with extrudable icing and soft cheese were used as printing materials to shape food structures (Southerland, Walters, and Huson 2011).

Other classification systems, such as traditional and non-traditional food materials (Feng, Zhang, and Bhandari 2019), or those based on chemical components, such as carbohydrates, fats, and proteins (Jiang et al. 2019; Perez et al. 2019; Portanguen et al. 2019), highlight different factors in printing materials, for example, their social acceptability or nutritional content. A review conducted by Voon et al. (2019) divided extrusion-based printing materials into two categories: natively extrudable printing materials (e.g., confectionery, dairy products, hydrogels) and non-natively extrudable printing materials (e.g., plants, meat). By contrast, this paper provides a comprehensive and systematic review of plant-based materials in extrusion-based food printing.

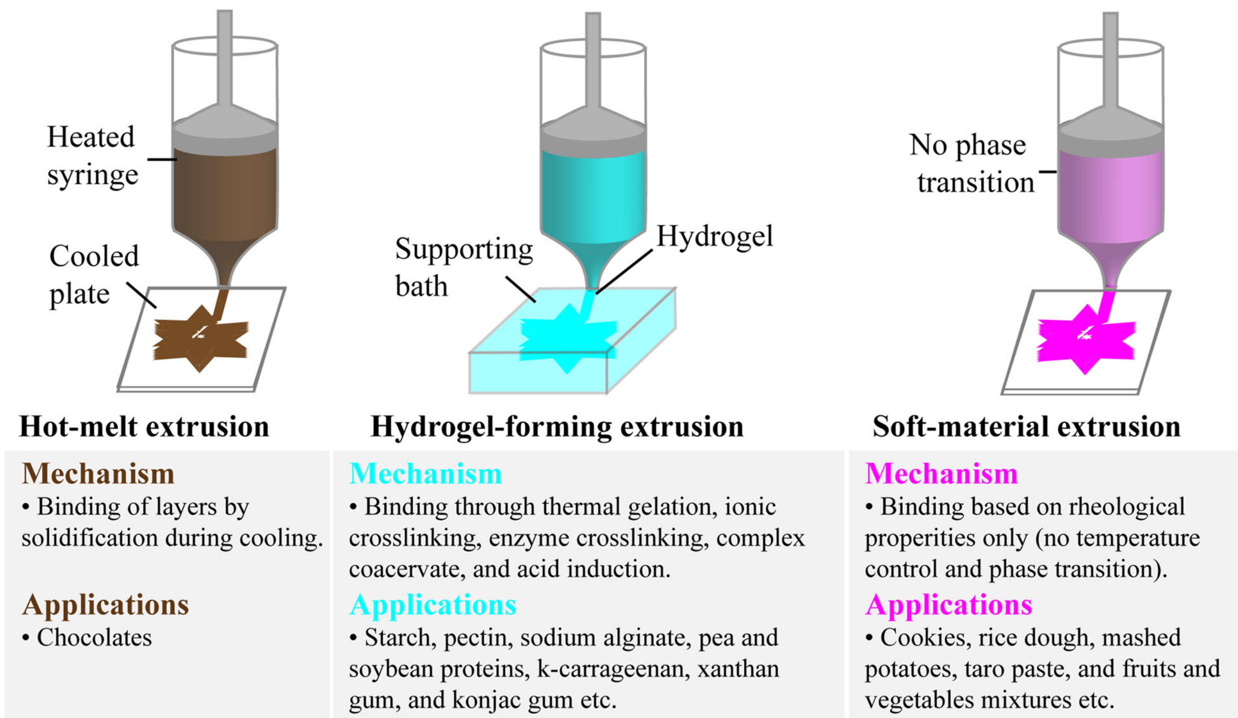

Figure 1. Binding mechanisms of extrusion-based food printing materials.

Binding mechanisms

Extrusion-based food printing can be divided into three categories according to the binding mechanisms of printing materials (see Figure 1): hot-melt extrusion, hydrogel-forming extrusion, and soft-material extrusion (Godoi, Prakash, and Bhandari 2016; He, Zhang, and Fang 2020; Sun et al. 2018).

In hot-melt extrusion, the melted semi-solid food polymer solidifies almost immediately after being extruded from the nozzle and welds to the previous layer (Sun et al. 2018). As such, it has been used to customize 3D-printed chocolate products (Hao et al. 2009, 2010; Mantihal et al. 2017). Here, controlling the temperature of the printing materials is key to ensuring printability.

Alternatively, hydrogel-forming extrusion is used to extrude or disperse hydrocolloid solutions into a polymer/hardening/gel supporting bath using a pipette, vibrating nozzle, or similar equipment (Sun et al. 2018). The rheological properties and formation mechanisms of printing materials are important factors affecting the extrusion of hydrogels. The printing materials should exhibit certain viscoelastic properties and be receptive to being transformed into self-supporting gels before successive layer deposition. To prevent the premature gelation of printing materials inside the nozzle, the gelation time must be controlled. Many hydrogel formation mechanisms have been identified, including thermal gelation, ionotropic crosslinking, enzyme crosslinking, complex coacervate formation, and acid induction (He, Zhang, and Fang 2020).

Conversely, soft-material extrusion is performed without temperature control and phase transition. It refers to the smooth extrusion of natively printable materials at room temperature (F. Yang, Zhang, Prakash, et al. 2018). Successful printing depends on the viscosity of the printing materials, which must be sufficiently low to allow extrusion through a thin nozzle while being sufficiently high to support the structure post-deposition (Lille et al. 2018). Provided they conform to food safety standards, thickeners or additives can be used to achieve the desired rheological properties.

Plant-based materials for extrusion-based food printing

The regular consumption of plant-based foods can help prevent certain major diseases, such as cancer and cardiovascular disease (Satija and Hu 2018). This section discusses the wide variety of plant-based foods that have been developed into printable materials according to their binding mechanisms: hot-melt materials, hydrogel materials, and soft materials.

Hot-melt materials

Chocolate is the principal hot-melt material used in extrusion-based food printing, with its use first demonstrated using the Fab@home manufacturing system by researchers at Cornell University (Periard et al. 2007). In 2010, Hao et al. (2010) founded Choc Edge Ltd and developed Choc ALM, the first chocolate 3D printer, a pioneering development in the commercialization of 3D chocolate printers. Subsequently, Choc Edge developed the Choc Creator, and several other companies developed their own 3D chocolate printers (see Table 1).

Chocolate is ideal for hot-melt extrusion because its main structural component is cocoa butter, which melts upon

Table 1. An overview of commercial extrusion-based 3D chocolate printers.

Company	Choc edge	Fouche chocolates	3D systems	3DCloud	Global 3D labs
Machine	Choc Creator	Fouche Chocolate Printer	CocoJet	QiaoKe	Chocobot
Country (city)	UK (Exeter)	South Africa (Centurion)	USA (Rock Hill)	China (Beijing)	India (Bangalore)
Year	2012	2014	2015	2015	2015

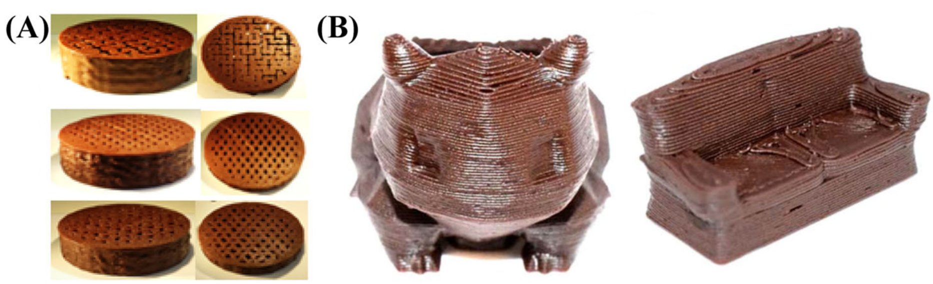

Figure 2. (A) 3D-printed plant sterol powder-enriched chocolate structures with an infill percentage of 60% Reprinted with permission from (Mantihal, Prakash, and Bhandari 2019a), copyright (2019), Elsevier; (B) Structures printed via direct chocolate-based ink 3D printing (Karyappa and Hashimoto 2019).

heating and solidifies after cooling to form a self-supporting layer (Jiang et al. 2019). It is paramount that the chocolate retains its structure during layer-by-layer deposition, with its "self-supporting" capability depending on thermal properties such as the glass transition temperature (T_g) and melting point, which are crucial in the post-deposition solidification of deposited layers (Mantihal et al. 2017). As the melting point increases, cocoa butter crystallizes into six different polymorphs, with this complex crystallization behavior dictating the thermal, rheological, and physical properties of the final product (self-supporting layer, gloss, and blooming during storage). However, this behavioral complexity can pose challenges for chocolate formulation (Marangoni and McGauley 2003). This was demonstrated by Hao et al. (2009), who found that different chocolate formulations (i.e., milk, white, and dark chocolates) showed distinct printing characteristics. In particular, changing the source and content of fat within the structure can cause changes in inter-particle interactions. Specifically, the addition of fat and lecithin can coat sugar and cocoa particles and reduce their interaction, resulting in significant reductions in the rheological, textural, and thermal parameters of the chocolate. Moreover, lower concentrations of cocoa butter corresponded to a higher ratio of solid particles, thereby promoting the interaction between solid particles and increasing the values of rheological properties.

From a materials perspective, significant research has been devoted to the use of additives in chocolate formulation. Not only can additives improve the nutritional properties of printed food, but can also enhance their rheological properties. Across several studies, it was found that the addition of magnesium stearate helped to improve fluidity during the deposition process, making the chocolate formulations more "printable." The effects of magnesium stearate powder (as a processing aid) and plant sterol powder (as a processing and nutritional aid) on the thermal, rheological, and tribological properties of chocolate were also investigated. Although the additives did not affect the thermal properties, they increased the friction coefficient of the samples, improved their fluidity, and reduced slipping during screw extrusion (Mantihal, Prakash, and Bhandari 2019a; Mantihal et al. 2017, 2019). Elsewhere, Hao et al. (2019) optimized the formulation of milk chocolate containing vitamin C, cranberry powder, and methylcellulose and further reported the need to limit the additive content to approximately 1% by weight, with excessive dry powder reducing the fat percentage and causing the chocolate to collapse.

In addition to the recipe, optimizing the printing process and parameters is crucial for successful chocolate printing. The key parameters affecting the geometric accuracy of chocolate deposition, including the nozzle aperture diameter, nozzle height, extrusion, and axial motion velocity, were identified by Hao et al. (2009, 2010) through a combination of theoretical and experimental analyses. Furthermore, the authors investigated the extrusion behavior of 3D-printed chocolate, revealing that the viscosity of chocolate was relatively stable for temperatures from 32–40 °C and pressures from 3.5 to 7.0 Pa, with the shaping of the chocolate becoming difficult at higher temperatures and pressures. The design of the printing structure is crucial when printing chocolate with complex structures, as highlighted by Mantihal et al. (2017), who designed chocolate structures with cross support, parallel support, and no support and studied the influence of different support structures on their snapping properties. The study revealed a strong correlation between the breaking strength and the chosen support structure, with cross supports proving more effective than parallel supports for creating a stable hexagonal structure. Mantihal, Prakash, and Bhandari (2019a) found that when the infill percentage was 60%, star-shaped and honeycomb infill patterns produced the toughest and most stable structures, as shown in Figure 2(A). Meanwhile, the authors conducted a sensory evaluation and consumer perception study using 3D-printed dark chocolate with improved texture, which showed that consumers preferred the appearance of samples with 25% and 50% infill percentages to those with 100% (Mantihal, Prakash, and Bhandari 2019b). These results highlight the influence of commercial factors on the application of 3D-printed chocolate.

The development of new printers and printing methods has been another focus of recent research. Lanaro et al.

(2017) constructed a melt extrusion 3D printer based on readily available open-source components and optimized several key manufacturing parameters, including variables such as the speed of the printer head, and the extrusion and cooling rates, enabling it to print complex 3D objects. In addition, Hao et al. (2019) improved the printing time and quality via algorithms for adaptive layer thickness and scanning speed. Although the hot-melt extrusion of chocolate is simple, the operating temperature needs to be controlled precisely over a very narrow range. To complement currently available 3D-printing methods for chocolate, Karyappa and Hashimoto (2019) mixed ready-made chocolate sirup with 5–25% w/w cocoa powder paste to prepare chocolate-based inks with desirable rheological properties. The researchers created complex 3D shapes using direct ink writing (DIW) 3D printers without temperature control, as shown in Figure 2(B), representing the first realization of direct printing using liquid chocolate-based ink, which they called chocolate-based ink 3D printing (Ci3DP). The simplicity and flexibility of Ci3DP promises considerable potential for fabricating complex chocolate products without requiring temperature control.

Despite the widespread commercialization of 3D chocolate printing, it is relatively underreported in the literature. The above examples provide directions for the development of chocolate-based printing materials, including the development of nutrient enrichment/texture-improving formulations, the optimization of printing processes and parameters, specialized printer design, and the creation of new printing methods.

Hydrogel materials

Many hydrophilic polymers such as polysaccharides, peptides, and proteins, also known as hydrocolloids, can be dispersed in water to form hydrogels (He, Zhang, and Fang 2020). In general, research on hydrogels for use in 3D printing involves one or more of the following: developing new hydrogel materials, optimizing printing parameters, developing or improving printers, and establishing theoretical models of the materials or printing processes.

Development of new hydrogel materials

Most studies aim to optimize formulations and determine the effects of different ingredients, ingredient concentrations, and other conditions on the rheology and printability of hydrogels, as well as to elucidate relationships between rheology and printability. For example, Gholamipour-Shirazi, Norton, and Mills (2019) designed printable pastes for extrusion-based food printing based on a series of hydrocolloids. The researchers found that the pastes were printable for phase angles and relaxation exponents ranging from 3–15° and 0.03–0.13, respectively. This may serve as a design principle for hydrocolloid-based food printing process, with the rheological measurements informing the development of novel 3D printing materials. In addition to single hydrocolloids, other complex materials are under consideration. For example, Vancauwenberghe et al. (2017) prepared a series of pectin gels using various materials (e.g., low methoxylated pectin, bovine serum albumin, and $CaCl_2$) and changing various parameters, including sirup concentration and stirring speed, and they realized the printing of customized water-based porous food simulants (see Figure 3(A)). Alternatively, García-Segovia et al. (2020) developed gel systems comprising sirup, xanthan gum, and konjac gum, while Liu, Bhandari, et al. (2019) developed multi-component gel systems composed of carrageenan-xanthan-starch.

Evidently, the research on polysaccharide hydrogels is extensive, with research on the use of vegetable protein and polysaccharide compounds also attracting attention. To date, pea and soybean proteins have been isolated and utilized to develop printing materials. For example, the effects of pea protein on the printability of potato starch-based 3D printing materials with respect to the particle, crystal, and chemical structures, as well as the textural and thermal

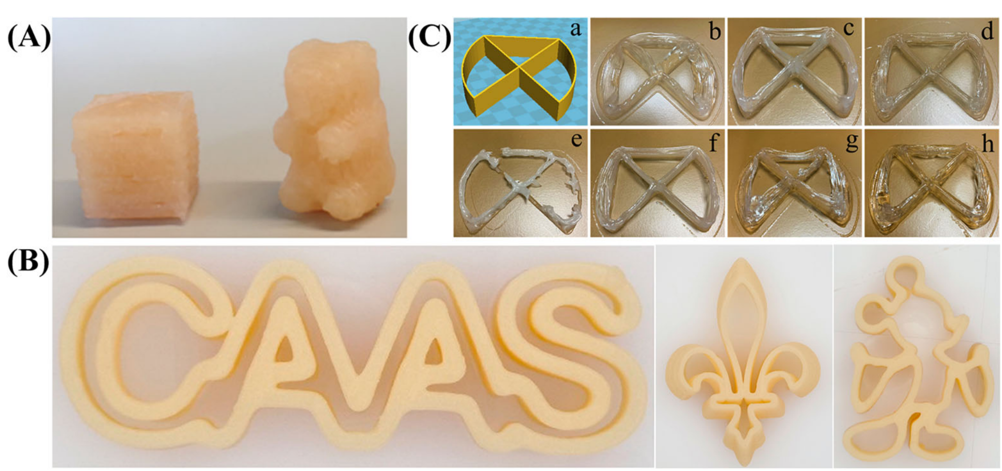

Figure 3. (A) 3D objects printed using pectin-based ink composed of 55 g/L LM pectin + 12.5 mM $CaCl_2$ + 25% (v/v) sugar sirup + 5 g/L BSA + edible colorant; (B) 3D geometries produced using an SPI mixture comprising 0.5% (w/v) sodium and 10% (w/v) gelatin (SPI: soy protein isolate); (C) Printed PS produced under different hot-extrusion printing temperatures and starch concentrations (a, model; b, 70 °C/10%; c, 70 °C/15%; d, 70 °C/20%; e, 60 °C/15%; f, 65 °C/15%; g, 75 °C/15%; h, 80 °C/15%) (PS: potato starch). Reprinted with permission from: Vancauwenberghe et al. (2017), copyright (2017), Elsevier; Chen, Mu, et al. (2019), copyright (2019), Elsevier; Liu, Chen, et al. (2020), copyright (2020), Elsevier.

properties, were investigated by Feng et al. (2018), revealing that a pea protein concentration of 1% delivered the best printing quality. Similarly, Chen, Mu, et al. (2019) showed that soy protein isolate (SPI) mixed with sodium alginate and gelatin produces ideal printing materials (see Figure 3(B)). These studies have provided a platform for the development and application of vegetable proteins in food printing, which, given their rich nutritional and functional properties, have significant potential for future 3D-printed food products.

Optimization of printing parameters

The optimization of common parameters (e.g., layer height, nozzle speed, nozzle height, and/or diameter (Diañez et al. 2019)) and printing temperature have been investigated to refine the rheological properties of hydrocolloids produced via thermal gelation. Notable studies include the discovery that starch suspensions (15–25% w/w) exhibit the best hot-extrusion properties and shape stability when heated at 70–85 °C (Chen, Xie, et al. 2019). Additionally, investigating the effects of starch concentration and printing temperature on hot-extrusion printing revealed that the temperature-dependent structural transformations in potato starch correspond to the rheological properties of hydrogels (see Figure 3(C); Liu, Chen, et al. 2020). The effects of printing temperature have also been studied regarding the uniformity and dimensional accuracy of 3D-printed gelatin-carrageenan gel systems (Warner, Norton, and Mills 2019).

Printer development and improvement

Unlike other printing materials, the mechanism of hydrogel formation limits its applicability using ordinary extrusion-based printers. To address this, some researchers have implemented printer modification to improve hydrogel printing efficiency. For example, Vancauwenberghe, Verboven, et al. (2018) designed a coaxial extrusion print-head for pectin-based food simulants, allowing food materials to flow internally while a $CaCl_2$ crosslinking solution flowed externally. Moreover, as gelation occurred during the printing process, no post-printing treatments were required. By varying the material composition, layer height, outflow velocity, and $CaCl_2$ concentration, suitable pectin-based printing materials were fabricated. Alternatively, Diañez et al. (2019) developed a forced convection system using ambient air to accelerate the cooling of the printing layer, thereby enabling in-situ gelatinization printing of k-carrageenan within a few minutes. However, this design requires further refinement to preserve shape fidelity. Ultimately, printer designs can be customized according to the characteristics of hydrogel materials.

Development of theoretical models

The effects of changing the parameters of printing materials and processes are often explored via computational simulations. For example, researchers have established a prediction model for pectin-based printing materials and obtained 3D-printed food simulants with predefined microstructures and textures (Vancauwenberghe et al. 2017; Vancauwenberghe, Verboven, et al. 2018). Analytical and finite element models were used to predict the textural properties of printed honeycomb structures, showing that the modeled textural properties decreased as the porosity increased, in agreement with empirical observations. The actual porosity of the printed structure yielded a good fit between the analytical model and the measured Young's modulus, with the validated finite element model providing a basis for designing additional complex structures (Vancauwenberghe, Delele, et al. 2018). Recently, the effects of material properties and process parameters on the velocity, shear rate, and pressure fields in the flow channel were analyzed (F. L. Yang, Guo, et al. 2019), leading to an improved 3D printing process for lemon juice-based gels. In summary, the use of models has great practical significance and provides useful reference data that inform printing processes and structural designs while reducing trial and error.

Soft materials

Cereal-based foods

Cereals, including wheat, corn, and rice, are characterized by their large starch content. Starch possesses good shear stability, with the starch molecule interactions that occur at high starch concentrations helping to maintain the stability of deposited structures (Feng, Zhang, and Bhandari 2019). Therefore, cereals have been used extensively in extrusion-based printing, including for pizza, cookies, and rice dough, and cereals are regarded as one of the most successful printing materials developed to date.

Influence of formula ingredients. Printable food materials typically comprise many components, whose proportions and physical properties can impact the printability and stability significantly. Lipton et al. (2010) developed a new cookie recipe suitable for 3D printing by changing the ratios of traditional cookie ingredients: increasing the relative butter content improved printability (at the expense of shape stability) while increasing the concentration of egg yolk improved the shape stability. Elsewhere, F. Yang, Zhang, Prakash, et al. (2018) found that sucrose, butter, and flour were all integral to the printing performance. Within a certain range, as the percentage of sucrose, butter, and flour increased, the gel strength, elasticity, and viscosity of the dough increased (and the ductility decreased), which helped improve their malleability and stability. However, above a certain threshold, the extrudability deteriorated, causing printing discontinuities. Similarly, Liu, Liang, et al. (2019) found that extrusion printing discontinuities occurred when the percentage of flour, water, olive oil, and freeze-dried mango powder exceeded a certain threshold. Therefore, deducing the optimal balance of ingredients in different food systems is an ongoing challenge and key to the successful implementation of 3D food printing.

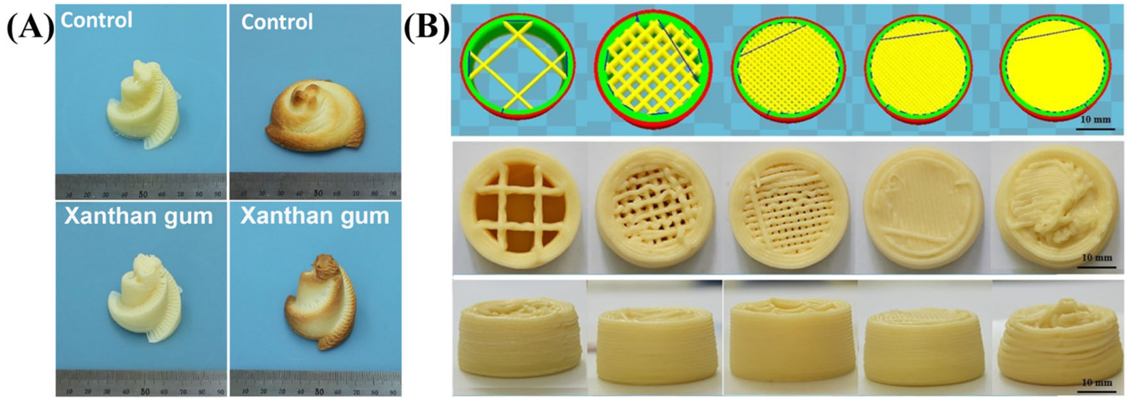

Figure 4. (A) 3D printed cookies (control vs. with xanthan gum) before and after baking; (B) Photos of 3D printed dough with filling ratios (from left to right) of 10%, 30%, 50%, 70%, and 100% (the top, middle, and bottom image rows show the 3D model, top view, and elevated side view, respectively). The needle diameter was 0.58 mm, the printing pressure was 600 kPa, and needle movement rate was 6 mm/s. Reprinted with permission from: Kim et al. (2019), copyright (2019), Elsevier; Liu, Liang, et al. (2019), copyright (2019), Elsevier.

Addition of additives. For a typical soft-material extrusion ink such as dough, the rheological properties are critical to successful printing. It is sometimes necessary to add additives to ink formulations to improve their rheological properties and enhance their printability, especially for grains with low starch content, which have low viscosity. Kim et al. (2019) discussed the effect of using hydrocolloids on the dimensional stability of 3D-printed cookie dough during post-processing. The results showed that cookie dough containing 0.5 g/100 g xanthan gum results in smooth printing as well as 3D shape preservation during post-processing, as shown in Figure 4(A). Additionally, Liu, Tang, et al. (2020) studied the feasibility of printing rice (waxy rice, japonica rice, and indica rice) pastes, as well as the effects of sodium alginate (SA) on their physicochemical and structural characteristics. Rice pastes containing SA demonstrated shear-thinning properties, indicating their suitability for 3D printing. The highest printability and accuracy were obtained for japonica rice with an SA concentration of 0.50%.

Influence of process parameters. Further to the influence of material composition on dough printability, several studies have reported the influence of printing parameters on the printing precision and texture of cereal-based foods, for example, the speed, diameter, and height of the nozzle, the extrusion speed, and the internal infill percentage (Anukiruthika, Moses, and Anandharamakrishnan 2020; Liu, Liang, et al. 2019; Yang, Zhang, Fang, et al. 2019). Liu, Liang, et al. (2019) ascertained the optimal compression pressure, needle speed, needle diameter, and internal filling rate (600 kPa, 6 mm/s, 0.58 mm, and 50%, respectively) required to produce 3D-printed dough with a well-organized packing structure, clear internal texture profile, and minimal deformation (see Figure 4(B)). Similarly, Severini, Derossi, and Azzollini (2016) analyzed the effects of infill density and height on the printing performance of wheat dough. The dough deposition became irregular as the layer height increased, with the fracture strength correlating closely to the infill level. For brown rice, Huang et al. evaluated the effects of nozzle size (0.84 mm, 1.20 mm, 1.56 mm), perimeters (3, 5, 7), and infill densities (15%, 45%, 75%) on the printing quality, showing that both the nozzle size and perimeter influenced the dimensional characteristics of 3D-printed samples, whereas the infill density did not; that is, the smaller the nozzle size, the better the dimensional performance. Moreover, textural properties (hardness and viscosity) correspond closely to infill density, followed by the perimeter and nozzle size. In addition to the voidage, the nozzle size also affects the textural properties indirectly by changing the number of layers deposited (Huang, Zhang, and Bhandari 2019).

Post-processing. Three-dimensionally printed dough usually requires post-processing such as baking, steaming, or frying. During post-processing, many chemical reactions occur, including the Maillard reaction, protein denaturation, moisture content reduction, and physical changes in color, volume, and texture (Sun et al. 2018). Severini, Derossi, and Azzollini (2016) studied the changes in size, shape, microstructure, and texture of wheat-based foods, observing that the diameter of the samples decreased after boiling because of the water removal effect. It is also important to maintain shape and texture during post-processing. For example, Kim et al. (2019) improved dimensional stability by adding xanthan gum to cookie dough, while Yang, Zhang, Fang, et al. (2019) improved the structure and shape of baked dough by rapidly freezing the dough at −65 °C for more than 10 min between printing and baking. Furthermore, some researchers have proposed alternative post-deposition cooking methods. For example, Sun et al. (2018) proposed immediate infrared radiation in place of baking. However, this has yet to be realized. Nevertheless, improving printing material formulations and developing new post-processing methods are evidently effective in addressing the undesirable changes incurred during the post-processing of dough, and they contribute valuable research insights.

Development of functional cereal-based food. From a nutritional perspective, the addition of innovative ingredients (e.g., edible insects or probiotics) to improve the nutritional benefits of cereal-based 3D-printed foods is an active

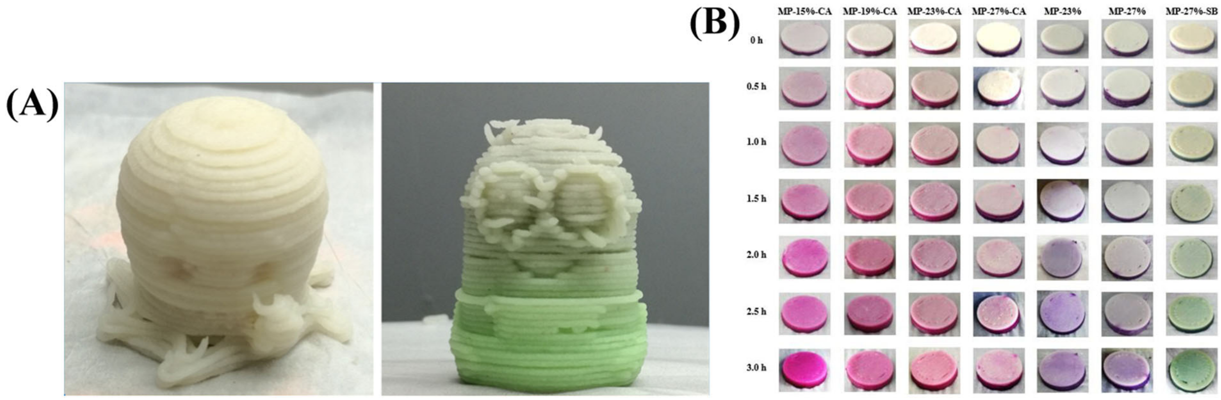

Figure 5. (A) Objects printed using KG-XG-MP (KG: k-carrageenan gum, XG: xanthan gum, MP: mashed potatoes). (B) Evolution of the post-printing color changes of MPs with different formulations. Reprinted with permission from: Liu, Zhang, and Bhandari (2018), copyright (2018), Elsevier; He, Zhang, and Guo (2020), copyright (2020), Elsevier.

research area. Recently, Severini, Azzollini, et al. (2018) developed wheat snacks that comprised wheat flour enriched with insect powder (yellow mealworm larvae), with the printed product providing greater amounts of protein and essential amino acids. Alternatively, Zhang, Lou, and Schutyser (2018) fortified dough with probiotics and studied their robustness to different printing structures and baking conditions. The final product, which possessed a honeycomb structure, met the definition of a probiotic food (number of viable bacteria > 10^6 CFU/g) after baking at 145 °C for 6 min. By adding *Arthrospira platensis* to cookie dough, Vieira et al. (2020) developed 3D-printed cookies with antioxidant activity. The cookies exhibited improved antioxidant activity and long-term color stability when the antioxidant extract was encapsulated into alginate microbeads as opposed to adding dried biomass or freeze-dried antioxidant extract. Studies such as these highlight the vast prospects of nutrient-enriched 3D-printed foods.

Fruit- and vegetable-based materials

As essential elements of human dietary requirements, fruits and vegetables provide carbohydrates, vitamins, minerals, antioxidants, and other bioactive substances (Li et al. 2017). The introduction of fruit and vegetable-based materials creates the opportunity to increase the availability and diversity of high-nutrition food-based printing materials.

Starchy vegetables. Similar to cereals, root vegetables such as potato, yam, and taro are rich in starch. As such, they have been widely adopted in 3D food printing (Feng, Zhang, and Bhandari 2019).

Mashed potatoes (MPs) are a prime example. They can be prepared directly from raw potato (Liu, Zhang, et al. 2018) or gelatinized potato flakes (Liu, Bhandari, et al. 2018). Although MPs themselves are extrudable, additives such as hydrocolloids are often added to maximize their printing performance. For example, Liu, Zhang, et al. (2018) found that MPs containing 2% starch are easily extrudable and provide a printed product with smooth shape, good resolution, and sufficient mechanical strength to support the deposited shape. Moreover, they demonstrated that gums, such as xanthan (XG), guar (GG), k-carrageenan (KG), and k-carrageenan-xanthan blends (KG-XG), can be used to tailor the rheological properties, microstructure, and 3D printing characteristics of MPs. Excluding XG, the addition of other gums can increase the viscosity, storage modulus (G'), and loss modulus (G'') of MPs. Printed objects comprising KG-XG-MP had a smooth surface structure due to the high printing precision (with the lowest dimensional deviation during printing), as shown in Figure 5(A). The authors postulated that the creaminess of XG yields a smooth surface structure, while the KG provides MP with sufficient mechanical strength for it to be self-supporting (Liu, Zhang, and Bhandari 2018). By contrast, Dankar, Pujola, et al. (2018) reported that increasing the concentration of glycerol and lecithin decreases the viscosity of MPs. The addition of agar or alginate generates additional convolution, thus increasing the stability of MP. Optimal printing performance and product stability were obtained using alginate and agar concentrations of 0.5–1.5% and 0.5–1%, respectively. Samples of MP supplemented with agar, alginate, butter, olive oil, and carrot all exhibited shear-thinning non-Newtonian behavior. Of these additives, butter forms lipid-starch complexes with starch molecules, maintains rigid network formation, and increases stability because of its high thixotropy and yield stress values (Dankar et al. 2019). Evidently, both the characteristics of individual materials and their interactions must be considered when selecting additives.

Improving the textural characteristics of 3D-printed food by manipulating the infill structure is a fledgling research direction. The infill structure of 3D-printed materials can significantly affect their mechanical strength and stability (Zhao et al. 2020). Liu, Bhandari, et al. (2018) printed MPs with various internal structures by varying the infill level (10%, 40%, 70%), infill pattern (rectilinear, honeycomb, Hilbert curve), and the number of shell perimeters (3, 5, 7). Additionally, the researchers developed 3D-printed air-fried potato snacks with unique internal structures and textural properties by changing the infill level and pattern (Liu, Dick, et al. 2020). By contrast, Feng et al. (2020) mixed potato processing by-products with yam and investigated the printing performance for infill levels of 20%, 50%, and

Table 2. Application of hydrocolloids in 3D printed fruit and vegetable-based materials.

Materials	Hydrocolloids	References
Mashed potatoes	Potato starch	(Liu, Zhang, et al. 2018)
Lemon juice	Potato starch	(Yang, Zhang, Bhandari, et al. 2018)
Lemon juice/ blueberry anthocyanin powder	Potato starch	(Ghazal, Zhang, and Liu 2019)
Mango juice concentrate	Potato starch	(Yang, Zhang, and Liu 2019)
Fruit-based snack (bananas, white beans, mushrooms, and lemon juice)	Pectin	(Derossi et al. 2018)
Lettuce leaf cells	Pectin	(Vancauwenberghe et al. 2019)
Fruit and vegetable blends (pears, carrots, kiwi, broccoli leaves, and avocados)	Fish collagen	(Severini, Derossi, et al. 2018)
Mashed potatoes	Pea protein	(Feng et al. 2018)
Purple sweet potato puree and mashed potatoes	Sodium alginate	(He, Zhang, and Guo 2020)
Spinach powder	Xanthan	(Lee et al. 2019)
Carrot callus	Alginate	(Park, Kim, and Park 2020)
Strawberry powder	Soybean protein isolate	(Fan et al. 2020)
Pumpkin and beetroot	Soybean protein isolate	(Phuhongsung, Zhang, and Devahastin 2020)
Papaya	Wheat starch	(Xu, Zhang, and Bhandari 2020)
Mashed potatoes/ strawberry juice concentrate dual extrusion	Xanthan and k-carrageenan blend/potato starch	(Liu, Zhang, and Yang 2018)
Mashed potatoes	Xanthan, guar gum, k-carrageenan, k-carrageenan, and xanthan blend	(Liu, Zhang, and Bhandari 2018)
Potato puree	Agar, alginate	(Dankar et al. 2019; Dankar, Pujola, et al. 2018)
Lemon juice	Potato starch, sweet potato starch, wheat starch, and corn starch	(Yang, Guo, et al. 2019)
Orange concentrate	Wheat potato, xanthan, guar gum, k-carrageenan, and gum arabic	(Azam et al. 2018)
Vegetable powders (broccoli, spinach, and carrot)	Hydroxypropyl methylcellulose, locust bean gum, xanthan, guar gum	(Kim, Lee, et al. 2018)

80% as well as printing structures with parallel, cross, and complex supports. The results showed that printing variables affect the physical characteristics (e.g., shape and hardness) of printed and air-fried products.

Food-based 3D printing is a field rich in innovation. For example, Liu, Zhang, and Yang (2018) used a dual extrusion 3D printer to separately print MP/strawberry juice gels and adjusted the textural properties by varying the infill percentage to produce multi-material structures with higher geometric complexity. Furthermore, He, Zhang, and Guo (2020) used a dual extrusion printer to produce colorful 4D-printed (3D printing which changes spontaneously over time) instant food. In this case, the pH-dependent color of anthocyanin was utilized to print MP/purple sweet potato puree exhibiting spontaneous color change (see Figure 5(B)). These results inspired further studies exploring the creation of colorful 3D structures and development of nutrient-rich, personalized 3D products by adding different plant powders. Liu, Bhandari, and Zhang (2020) mixed MPs with probiotics to study the effect of printing variables (nozzle diameter and printing temperature) on probiotic survival. Only the smallest nozzle diameter (0.6 mm) caused a decrease in probiotic viability from 9.93 log CFU/g to 9.74 log CFU/g. When MPs were stored in the printing nozzle bucket at 55 °C for 45 min, the number of live probiotics decreased more substantially (from 10.07 log CFU/g to 7.99 log CFU/g). In contrast, storing the samples at 5 °C for 12 days had no notable effect on probiotic activity.

Aside from MPs, a recent study by Huang et al. (2020) focused on the development of taro paste and the printing performances attainable using different additives (SA, sodium carboxymethyl cellulose, xanthan gum, guar gum and whey protein). The additives were shown to affect the texture and moisture distribution, and improve the apparent viscosity, storage modulus G′, and loss modulus G″. Samples incorporating SA demonstrated the best printing performance. Developing novel materials and enhancing the collective understanding of the mechanisms underlying components interactions are important objectives for future research.

Other fruit- and vegetable-based materials. Other than starchy vegetables, most fruit and vegetable-based materials have high water content and low viscosity, rendering them too fluid for printing. Therefore, printability must be prioritized. Except for dehydrating materials in a centrifuge (Zhu et al. 2019), adding hydrocolloids is the most common approach for improving the rheological properties. Table 2 lists the applications of hydrocolloids in 3D-printed fruit and vegetable-based materials. Among them, starches (especially potato starch) are most popular. For example, lemon juice gels containing 15% potato starch provide the smoothest surface and target-shape match without suffering compression deformation (see Figure 6(A); Yang, Zhang, Bhandari, et al. 2018). In addition to potato starch, wheat starch has been combined with orange concentrate and papaya (Azam et al. 2018; Xu, Zhang, and Bhandari 2020). Furthermore, Yang, Guo, et al. (2019) showed that the rheological and textural properties of lemon juice gels changed when supplemented with different starches. Gels containing potato starch exhibited favorable flowability because the amount of bound water was relatively low. Alternatively, gels containing wheat, sweet potato, and corn starches demonstrated heightened viscosity, heightened elasticity and hardness, and high cohesion alongside a tendency to agglomerate and age, respectively. Evidently, the printing performance of fruit and vegetable-based materials can be improved by adding different starches. In addition, Azam et al. (2018) proved that the gums (gum arabic, guar, k-

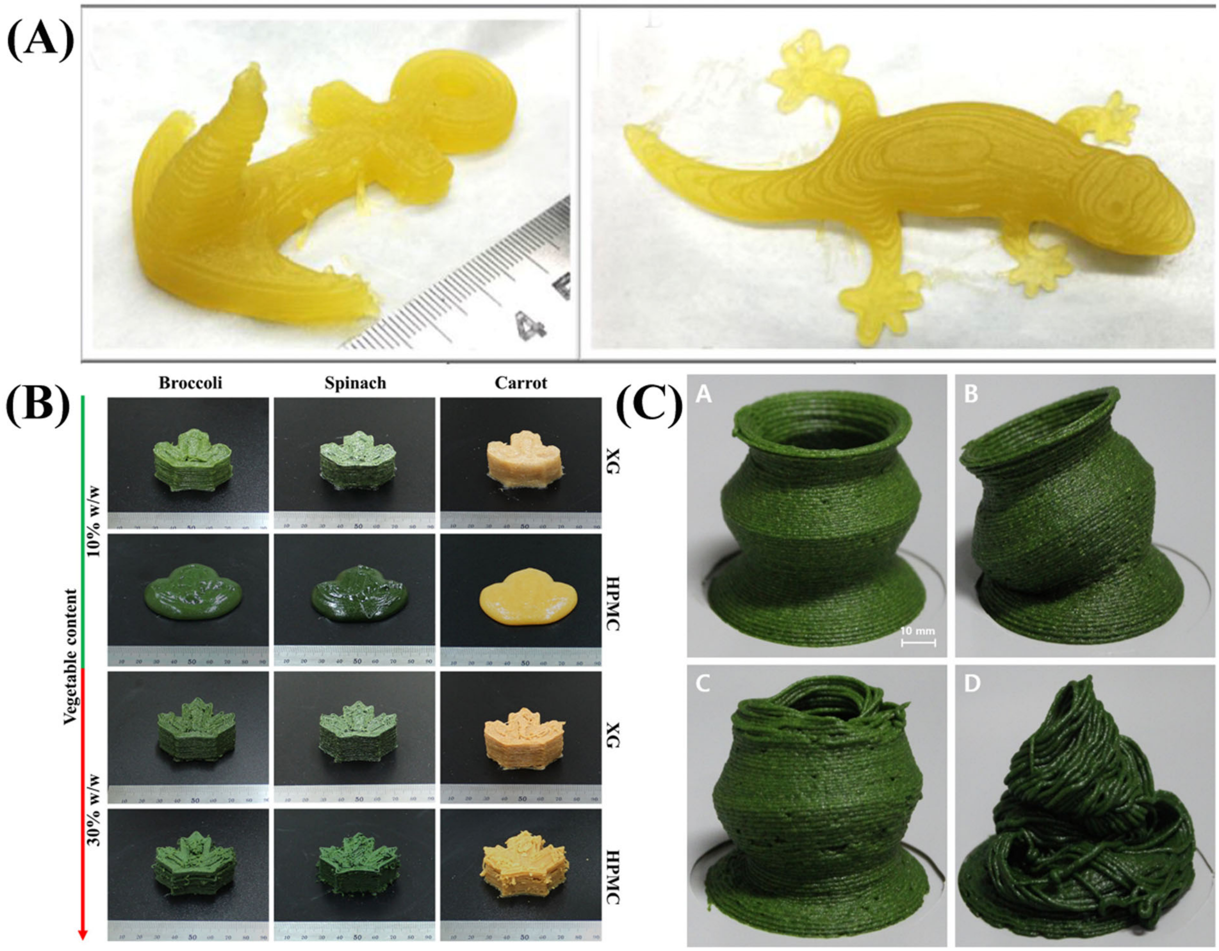

Figure 6. (A) 3D-printed products made with lemon juice gel and potato starch; (B) Images of XG- and HPMC-based products with vegetable powder contents of 10% and 30% (XG: xanthan gum, HPMC: hydroxypropyl methyl cellulose). (C) Images of 3D-printed spinach dispersions with particle sizes of A = 307 μm, B = 259 μm, C = 172 μm, and D = 50 μm. Reprinted with permission from: Yang, Zhang, Bhandari, et al. (2018), copyright (2018), Elsevier; Kim, Lee, et al. (2018), copyright (2018), Wiley; Lee et al. (2019), copyright (2019), Elsevier.

carrageenan, and xanthan) affect the rheology and 3D printing performance of mixtures combining orange concentrate and wheat starch.

In addition to concentrated pulp or juice, powdered fruit and vegetable extracts can be used as additives. The particle source, content, and size all impact the printability and rheological properties. Kim, Lee, et al. (2018) added vegetable powders (broccoli, spinach, and carrot) to hydrocolloid substrates. The printability and rheological properties of the hydrocolloids varied slightly for different vegetable powders (powder content of 10%). When the powder content increased to 30%, hydroxypropyl methylcellulose (HPMC), which has the lowest hydration, showed the greatest difference in rheology and printability, while xanthan gum (XG), which has high hydration, can restrain particle swelling, thus minimizing the rheological value of powders under high volume fractions and reducing the difference in printability between different vegetable powders (see Figure 6(B)). In addition, Lee et al. (2019) revealed that increasing the size of particles in spinach powder increased the storage and loss moduli, effectively improving the mechanical strength (see Figure 6(C)). Such studies aim to determine the parameters involved in preparing printing materials, such as milling time.

In addition to printability, the nutritional and sensory qualities of fruit and vegetable-based materials are also considered. For example, Derossi et al. (2018) designed an innovative fruit-based 3D snack for children aged 3–10 years. Bananas, white beans, mushrooms, skimmed milk, and lemon juice were added to pectin solution to make complex printing materials containing vitamin D (0.75–1.5 mg/day), iron (0.58–1.2 mg/day), and calcium (49–97.5 mg/day), providing children with 5–10% of their recommended daily intake of these substances. Vitamin D-rich foods can also be obtained by printing a blend of wheat starch and orange concentrate (Azam et al. 2018). Interestingly, Severini, Derossi, et al. (2018) mixed a variety of fruits and vegetables (pears, carrots, kiwis, broccoli, and avocado) to produce a "printable smoothie," with a pyramid shape designed to attract consumers. Nevertheless, the number of fruit and vegetable-based materials currently being used in 3D printing remains small. Therefore, there is a huge demand for the development of new fruit and vegetable-based printing materials with good printing performance, rich nutrition, and high sensory quality.

Printing parameter optimization is also essential to improve the printing precision of fruit and vegetable-based materials (Derossi et al. 2018; Severini, Derossi, et al. 2018;

Yang, Zhang, Bhandari, et al. 2018). The pre- and post-treatment of materials impact the printing performance considerably (He, Zhang, and Fang 2020). Yang, Zhang, and Liu (2019) studied the effect of microwave vacuum drying (MVD) post-treatment on mango juice gels, comparing the flavor, texture, moisture distribution, and dielectric properties of the printed products at different drying times (0, 2, and 4 min). Three-dimensionally printed products subjected to 4 min of MVD post-treatment exhibited the best shape retention and accuracy. Elsewhere, Xu, Zhang, and Bhandari (2020) investigated the effect of pre-treating materials using a combination of microwaves and ultrasound on the 3D printing quality of wheat starch-papaya systems. Specifically, the effects of using low microwave powers (60 W, 70 W, and 80 W) on gelatinization, rheological properties, dielectric properties, moisture distribution, texture, microstructure, and printing accuracy were compared. The results showed that the samples pretreated with 80-W microwaves exhibited the best support stability, line uniformity, and height retention. Additionally, Fan et al. (2020) studied the influence of microwave-salt synergetic pretreatment on the 3D printing performance and self-supporting behavior of soybean protein isolate-strawberry inks, demonstrating that salt and microwave treatments can significantly improve the printing accuracy and self-supporting performance of such inks.

In addition, Ghazal, Zhang, and Liu (2019) prepared attractively colored, healthy 4D foods using blueberry as a printing material. As the most colorful food materials, fruit and vegetable-based materials provide exciting opportunities for colorful 4D food printing. Moreover, Vancauwenberghe et al. (2019) encapsulated lettuce leaf cells into pectin-based bioinks to create innovative edible printing materials resembling plant tissues, while Park, Kim, and Park (2020) prepared callus-based bioinks by mixing alginate with a carrot callus dispersion. These studies emphasize the possibility of imitating fruits and vegetables using 3D printing technology.

Challenges and prospects

3D printing technology has developed rapidly in food-based industries in recent years. However, the widespread application of 3D food printing still faces many challenges, several of which are discussed herein. In addition, we propose possible solutions and turn our attention to emerging trends that are set to define the future of research on 3D food printing.

Printing precision and shape stability are the biggest challenges for 3D printing technology to overcome. To achieve accurate printing, further understanding of the relationship between the properties of food materials and their printability is required to develop new printing materials that deliver better performance. Combining 3D food printing with other emerging technologies may be an effective approach. For example, microwave and ultrasonic technologies are used for pretreatment or post-processing to improve the printing precision and shape stability of materials (Fan et al. 2020; Xu, Zhang, and Bhandari 2020). However, we emphasize that processing technologies should not be used to the detriment of the nutritional value and sensory quality of food materials but should minimize nutritional losses and sensory changes in tandem with improving printing performance. Alternatively, promising natural biopolymers can be modified to deliver materials with enhanced printability. For example, Maniglia et al. (2019, 2020) observed that ozone-modified and dry heat treatment (DHT) technology-modified cassava starch both offer better printability than natural starch.

At present, 3D-printed food typically satisfies a single requirement, for example, nutrition, color, flavor, and texture. Moving forward, 3D food printing technology has the potential to expand product functionality and create personalized nutritional foods. For example, printing customized soft food to facilitate chewing and swallowing for the elderly, while simultaneously addressing problems such as vitamin D deficiency (Azam et al. 2018; Zawada et al. 2018). Thus, mixing multiple materials can compensate for the nutritional deficiencies of a single material. Material sustainability is a further consideration; the use of renewable foods such as edible algae and insects to produce 3D-printed food is gaining support (Severini, Azzollini, et al. 2018), while developing effective uses for food processing by-products can reduce food waste. According to the data published by the UN's Food and Agriculture Organization, one-third of all food produced globally for human consumption is wasted, equating to 1.3 billion tons annually, of which 20% is meat and dairy products, 30% is cereals, and 45% is fruits and vegetables (Portanguen et al. 2019). These by-products contain proteins, dietary fibers, polyphenols, vitamins, and minerals, which possess high nutritional value. Therefore, combining food processing by-products with 3D printing technology to form a sustainable food production system would have an enormous impact from a sustainability perspective.

Beyond nutrition, the color, flavor, and texture of food are crucial elements of the eating experience, adding to the number of factors that 3D printing must replicate to achieve commercial success. The internal structure of 3D-printed objects can be changed by changing the filling pattern and percentage, thereby controlling material density and constructing novel foods with diverse microstructures and textures. Moreover, 4D printing will gain increasing popularity. Phuhongsung, Zhang, and Bhandari (2020) used a formulation comprising soybean protein isolate, K-carrageenan, and vanilla flavoring, which was then microwave-heated, to obtain 4D-printed products exhibiting automatic flavor change. In addition, the use of smart materials in 3D printing raises the possibility of products that can transform themselves instantaneously when exposed to predetermined stimuli, such as temperature, air humidity, ultraviolet light, electric, magnetic field, or pH, to form new structures with different shapes, colors, or flavors. Such products have immense potential for food processing applications (Miao et al. 2017; Shin, Kim, and Kim 2017).

Production efficiency is another challenge facing commercial 3D food printing. Although the printing speed or nozzle diameter can be increased, this often results in decreased printing precision and resolution, and it should be adopted only if the printing precision can be preserved. Alternatively, researchers have proposed increasing the printing speed via adaptive algorithms, which could adjust the printing parameters to balance the printing quality and time (Voon et al. 2019). Another potential approach involves using multi-nozzle printers to manufacture multiple objects simultaneously. To date, no industrial-scale printers offer this function, and the development of low-cost, highly reliable printers providing fast production remains a challenge (Nachal et al. 2019). Accordingly, further research to improve production efficiency and printing precision is required to accommodate the inevitable increase in the complexity of the control systems for such printing devices.

Ensuring the safety and longevity of 3D-printed foods is also a major challenge, as most 3D-printed foods have a limited shelf life. For example, the structural rheology of 3D-printed puree or dough changes two hours after production (Lipton et al. 2015), while certain fruit and vegetable-based 3D-printed products have been found to support high concentrations of microorganisms during storage in air, highlighting potential food safety issues (Severini, Derossi, et al. 2018). Therefore, ensuring that 3D printing processes conform to rigorous hygiene and quality regulations is essential before they can be applied in restaurants and factories.

Conclusion

This paper provides a systematic review of the applications of plant-based materials in extrusion-based food printing. The hot-melt, hydrogel, and soft (cereal- and fruit/vegetable-based) materials used in extrusion-based food printing should meet three essential requirements: printability, applicability, and post-processing suitability. Current research is mostly focused on the development of new plant-based printing formulations that deliver better performance, including the addition of nutritional and functional ingredients to supplement traditional materials such as chocolate and hydrogels, or the incorporation of additives (e.g., hydrocolloids) to improve the material printability, particularly natively non-printable materials such as fruits and vegetables. Meanwhile, further to the optimization of printing process parameters, recent research is displaying a trend toward the design of new printing structures that improve the customizability and target-specificity of 3D-printed foods. In addition, the development of specialized printers and new printing methods, as well as computational models and algorithms, can improve printing efficiency, and it requires in-depth study in the future.

Yet, 3D food printing technology still faces many challenges regarding food quality and printing processes. From a materials perspective, the development of nutrient-rich materials with strong printability is paramount, while food processing by-products can be utilized to improve ingredient sustainability. Moreover, new technologies for pre- and post-treatment offer great scope to enhance the printing performance of materials. Finally, 4D printing also promises tremendous application potential. The development of materials with transformable shapes, colors, and flavors will generate huge interest in printed food and promote the commercialization of 3D food printing technology.

Declaration of interests

There are no conflicts to declare.

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