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Advances in plant-based raw materials for food 3D printing

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A B S T R A C T

Three-dimensional (3D) printing is an emerging technology in the food industry, and in recent years, with its unique advantages, it has been widely used in the food industry. Its organic combination with food production provides customization, personalization, and intelligent features. The combination of plant-based food raw materials with 3D printing technology to produce food has a broad space for development in catering to people's pursuit of healthy diets. To facilitate a more comprehensive understanding of the application of plant-based food raw materials in food 3D printing, we classified inks into molten, soft, and hydrogel materials according to their state of existence and rheological properties under different conditions. The applications of different plant-based raw food materials in 3D printing are reviewed separately by ink type. This is expected to enrich the coverage of the review in related fields, enabling a quick understanding of the research direction and enriching the research content.

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1. Introduction

Three-dimensional (3D) printing technology, also known as additive manufacturing, is a promising technology that uses a computer-controlled system to control the layer-by-layer stacking of raw materials to obtain the target object[1]. Fig. 1 provides an overview illustration of the current status of the application of 3D printing technology in various fields. Owing to its low cost, fast preparation, ease of use, and shape-shaping, it is widely used in materials, machinery, food manufacturing, medicine, and other fields[2-3]. Especially in the food manufacturing sector, the use of different ink types and printing methods makes the technology a perfect fit for the diverse and customized food products of the future. With the development of the food industry, people's requirements for food are becoming increasingly diversified and are no longer restricted to the belly, but also consider the health, customization, and functionalization of food[4]. Therefore, the organic combination of food production and 3D printing technology is suitable for future food development and has been developing rapidly in recent years.

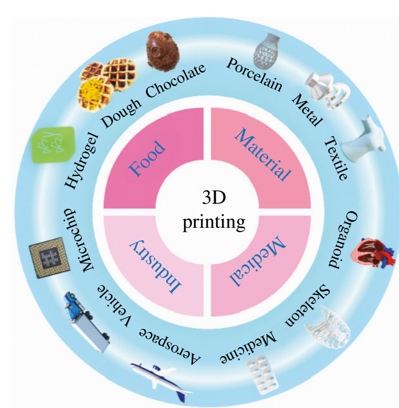

Fig. 1 Application of 3D printing in various fields.

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Extrusion-based printing (EP) is the most common food 3D printing technology used to build 3D objects by extruding print materials from a print nozzle and allowing them to be stacked layer by layer. The printing process can be divided into 4 stages: 1) ink preparation, 2) ink extrusion, 3) ink deposition, and 4) formation of a 3D structure[5]. Because no binder is used for binding in the printing process and the powdered material cannot be extruded directly, only the rheological properties of the raw material can be used to shape the object, which requires certain rheological properties of the raw material. In general, raw materials are considered printable when they meet the following conditions: they must be continuously extrudable and the extruded material must have the strength to support the 3D structure that needs to be established[5-6]. Thus, the selection and preparation of raw materials are decisive factors affecting the indicators of the final product.

Plant-based food materials have always been a sought-after object for a healthy diet because of their rich nutrient content, including fibers, polyphenols, and vitamins, and their ability to prevent cardiovascular diseases. The diversity of components among different plant-based raw food materials provides a wide scope of development for the preparation of 3D printing inks[7-8], which have been widely used in different 3D printing technologies. However, we have not seen a proliferation of 3D printed plant-based foods on the market because the printability of plant-based inks has been a constraint on the development of 3D printed foods. The plant-based ink system is a heterogeneous system characterized by the diverse properties of various plant components and the differentiation of specific target foods. Categorizing inks solely based on their composition fails to establish a distinct differentiation, as the relationship between ink composition and print quality necessitates a comprehensive evaluation of the ink's state of existence and rheological properties. Therefore, this paper classifies our common food systems into three main categories according to their state of existence and rheological properties: molten materials, soft materials, and gel materials to establish a more intuitive link between the ink and the quality of the prints. The grouping of inks of the same form of existence into one category also allows for more targeted advice and suggestions to improve the printability of vegetable-based inks and serves as a better source of inspiration. The current status of printing different plant-based raw food materials in different ink states and methods to improve printability are reviewed, providing a more intuitive and comprehensive summary of the study. Finally, the prospects and challenges of food 3D printing technology are discussed.

2. Molten materials

2.1 Chocolate

Molten materials, including fat-based foods, mainly rely on heating to change their rheological properties to make them printable[9]. The most common plant-based food ingredient used in food 3D printing for preparing molten materials is chocolate[10], as shown in Fig. 2. Chocolate is composed mainly of cocoa butter, and the complex fat composition and microscopic interparticle interactions in chocolate confer its complex rheological properties. The rheology of chocolate is mainly influenced by its solid content, particle size, fat content, moisture, and emulsifiers. Some scholars have investigated the effect of the solid content of chocolate on its 3D printing quality and prepared chocolate inks with different solid contents by controlling the addition of cocoa powder and then printing them separately. They reported that the best print quality is achieved when cocoa powder is added at 20% (*m*/*m*). When the amount added is too low, a stable support structure cannot be formed due to the low solids content, while if the amount added is too high, the ink cannot be continuously extruded and 3D printing is not successful; an example of the relevant print is shown in Fig. 2A[11].

Fig. 2 (A) Chocolate ganache prepared by 3D printing under different cocoa powder (CP) additions (samples with 0%, 10%, 20%, and 30% (*m*/*m*) CP were labeled CP0, CP10, CP20, and CP30, respectively)[11]. (B) Low-fat 3D-printed chocolates using water-in-oil emulsions[17]. (C) Complex chocolate structures are printed by an optimized printing platform[20].

In addition to the solid content, the size of the particles in the chocolate ink also affects the printability of the chocolate and the texture of the printed product. A larger particle size will make the chocolate taste gritty, while a smaller particle size will make the chocolate taste more delicate but will increase its viscosity and reduce its fluidity[12]. Thus, it is necessary to consider the appropriate particle size when designing the ink to ensure the quality of the product while simultaneously considering its printability. Do et al.[13] and Deou et al.[14] prepared low-fat chocolates with a low target viscosity without increasing the fat content of chocolate by optimizing the particle size distribution. However, most of these studies reflect the influence of the particle size index on the quality of the printed chocolate from a theoretical perspective, and a few studies have used particle size optimization to improve the rheology of chocolate 3D printing inks and use them for 3D printing; therefore, this aspect should be explored in future studies.

Another important factor affecting the rheological properties of chocolate is its fat content. The cocoa butter and other fats in chocolate cover the surface of the solid particles, lubricating and conferring the chocolate with good fluidity. These fats also affect the melting point and crystallinity of chocolate. Therefore, the viscosity and melting point of chocolate can be adjusted by changing the fat content. Afoakwa et al.[15] examined the effect of fat content on the rheological properties of chocolate in detail and found that the viscosity of chocolate decreases with increasing fat content; this is due to an increase in the particle size and a significant decrease in the specific surface area of the chocolate caused by an increase in the fat content. However, because fats such as cacao butter are expensive, their use to adjust the rheology of chocolate leads to higher production costs. In addition, their high saturated fatty acid content is thought to be associated with the development of diseases such as hypertension and insulin resistance[16]. Therefore, the use of emulsifiers and surfactants to adjust the rheological properties of chocolate has received widespread attention because they play a similar role to cocoa butter lubrication, can change the rheological properties of chocolate to improve printability, and can be adapted to the current needs of people's low-fat diets. You et al.[17] developed a low-fat 3D-printed chocolate using a water/oil emulsion based on Arabic gum instead of cocoa butter to regulate the rheology of the chocolate ink, as shown in Fig. 2B. Prosapio et al.[18] and Tirgarian et al.[19] used different emulsions instead of cocoa butter to adjust the rheology of chocolate, and both studies confirmed the feasibility of using emulsions to improve the printability of chocolate inks.

In addition to the properties of the material itself, the setting of print parameters is also an important factor in the final chocolate prints. In their study on the impact of printing parameters on chocolate print quality, Lanaro et al.[20] discovered that the distance covered by chocolate filaments remained unaffected by changes in the printing speed ranging from 300 to 700 mm/min. However, they observed that reducing the ambient temperature could enhance the distance covered. This finding holds significant relevance for the industrialized production of 3D printed chocolates, as it enables accelerated printing speed and improved production efficiency while ensuring high print quality. Liang et al.[21] also explored the effect of printing process parameters on chocolate printing results and successfully used existing commercial printers to improve the print quality and molding efficiency of chocolate, all of which have contributed to the popularity of 3D printed chocolate (Fig. 2C).

2.2 *Sugar*

Sugars, including sucrose, glucose, and fructose, are plant-based foods naturally produced in several fruit-bearing plants and are refined by a series of processes[22]. In modern society, it plays an important role in the diet and is not only recognized for its flavor and special sweetness but also plays a great role in food preservation[23]. The application of sugar in food 3D printing is attributed to its unique melting properties, allowing it to melt by heat or dissolve and consequently fuse with adjacent sugar particles to form the target structure.

The print quality and associated print parameters of sugar are affected by its purity and moisture content, and variations in these two conditions affect its melting point, glass transition temperature (T_g), and rheological properties[24]. Kim et al.[25] explored the effect of sugar purity on its printing performance in 3D hot extrusion by adding potato starch to sugar; they found that the sugar with 50% potato starch showed the best printability. The effect of the moisture content on the printability of sugar is mainly reflected in changes in its T_g and viscosity. The T_g is the critical temperature at which sugar is transformed into a viscoelastic state. Viscosity affects the fluidity of sugar after melting, both of which are important parameters for measuring sugar printability. Wang et al.[26] analyzed the rheology of caramel at different moisture contents and found that an increase in moisture content causes a decrease in the T_g and viscosity of the caramel, which may cause changes in the printing parameters and a decrease in the resolution of the printed product.

Based on the aforementioned studies, the significance of temperature in the 3D printing of sugar becomes evident. While it is possible to alter the melting temperature and T_g by adjusting the printing ink composition, careful temperature control throughout the printing process remains crucial. This is particularly critical when laser sintering is employed for printing icing sugar. Excessively high temperatures can result in sugar decomposition or excessive Maillard reactions with impurities present in the icing sugar, which could potentially generate carcinogenic substances and pose risks to both food safety and flavor.

The above review of previous studies provides references for the future development of sugar-based printing materials and the optimization of printing processes and parameters.

3. Soft materials

3.1 *Cereal-based raw materials*

The rheological properties of plant-based soft materials under shear conditions during and after extrusion are suitable for preparing inks for extrusion printing[27]. Currently, the common plant-based viscoelastic materials used for food 3D printing are prepared from cereals. Cereals are one of the most common raw food materials in our daily lives and are nutrient-rich, providing the body with nutrients such as lipids, carbohydrates, proteins, minerals, and vitamins. The consumption of whole grains also reduces the risk of cardiovascular diseases, hypertension, and cancer to some extent[28]. Cereal-based raw materials, which can be processed into food printing inks with excellent rheology owing to their rich starch content, are one of the most potentially developed food 3D printing raw materials[10]. The relevant 3D-printed food products prepared using cereal ingredients are shown in Fig. 3. The most widely used cereal-based food 3D printing material is dough, which is a solid but viscous foam-like mixture of flour, water, and yeast that is mixed and kneaded[29]. Dough exhibits shear thinning behavior during extrusion and maintains excellent support properties after extrusion[1,10].

The relationship between rheology and dough printability has been extensively studied for improved use in food 3D printing. The rheology of dough is mainly influenced by the starch and gluten content in the dough, the degree of starch pasting, and additives. Dough can be considered as a mixture composed of starch and gluten, both of which have different properties in the dough. Gluten exhibits rubber-like viscoelasticity and influences the mechanical properties of the dough, whereas starch is essential for the dough to exhibit shear thinning behavior. The interaction between the two in the

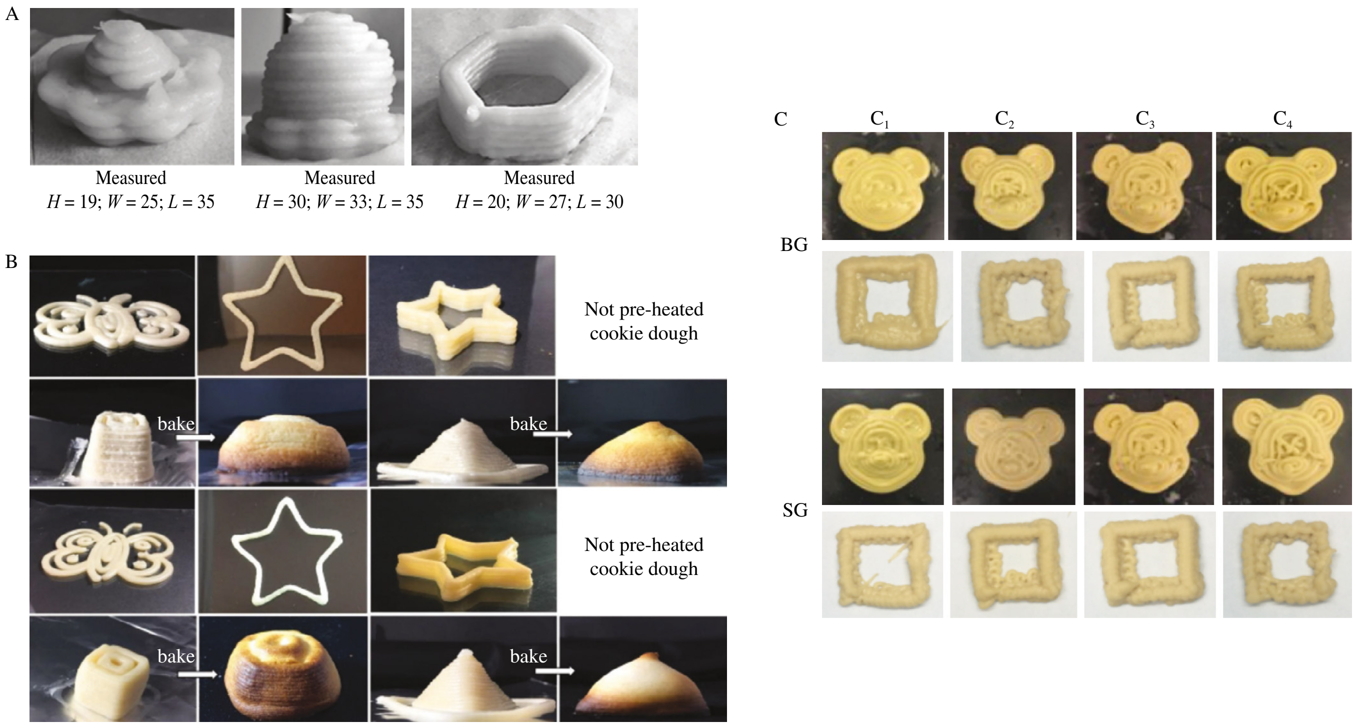


Fig. 3 (A) Heat-treated wheat dough for 3D printing[27]. (B) The effect of preheating on the print quality of biscuit dough[36]. (C) Effect of different butter additions (BG) and sucrose additions (SG) on the print quality of the dough (samples C1-C4 correspond to a fixed SG level of 6.6 g/100 g, with BG levels of 0, 3, 6, and 9 g/100 g, respectively. Additionally, another set of samples was prepared with a fixed BG level of 6 g/100 g and varying SG levels of 3.3, 5, 6.6, and 8.2 g/100 g, respectively).[37].

dough influences its rheology and printability[30-31]. Masbernat et al.[27] constructed wheat dough using a heat-treatment process and investigated the printability of the dough and the interactions between gluten and starch during processing. They revealed that after the heat treatment of the dough, the starch swells and the protein denatures and aggregates, with the two crosslinking each other to form a tight sticky network structure. This gives the dough good printability and mechanical properties, ensuring its successful printing; the printing results are shown in Fig. 3A. Song et al.[32] studied the effect of gluten content on the rheological properties of dough and found that an increase in gluten content can make the dough more elastic and robust, which is beneficial for improving the printing resolution and stability of printed products. The effect of starch content on the rheology of dough was also measured. It was found that increasing the starch content increased the dough viscosity and decreased the overall pasting temperature of the dough. This is in contrast to gluten because the increase in starch content provided an advantage in competitive hydration with gluten[33].

As mentioned above, the influence of dough composition on dough rheology and the degree of pre-gelatinization of starch, one of the main components of dough, also has a great influence on dough rheology and printability. Gu et al.[34] explored the effect of oat starch with different degrees of pasting on the rheological properties of oat dough. They found that the storage modulus (G') and loss modulus (G'') of dough kneaded at 25 °C, with oat starch pasting in the range of 15%–50%, increased with increasing pasting degree. However, both moduli decreased when the pasting degree was in the range of 50%–90%. This indicates that over pasted starch may destroy the elasticity and dense structure of the dough, which is not conducive for preparing 3D printing inks. The effect of starch pre-gelatinization on potato starch-gluten model dough was also studied by Xu et al.[35]. They found that the linear viscoelastic region of potato starch-gluten model dough increased significantly with increasing starch gluing within a certain gluing range, with a consequent decrease in tan δ. This suggests that higher starch gluing confers a more elastic structure to the dough. Pulatsu et al.[36] studied the effect of preheating on the rheology and printability of cookie dough prepared from a mixture of flour. They performed a detailed analysis of the complex interactions among starch pasting, protein aggregation, and fat release, whose combined effects imparted different rheologies and printabilities to the cookie dough under different processing conditions. The printability results are shown in Fig. 3B. The above results suggest that doughs with different degrees of pasting have different rheological properties, but the variation in this trend can be influenced by specific dough components. Thus, when using the degree of starch pasting to modulate dough rheology, attention should be paid to the actual application scenarios. The effect of the degree of starch pasting on the rheology and printability of the dough used to print different foods may vary depending on dough formulation and processing conditions (Fig. 3C).

3.2 Starch-based fruits and vegetables

Fruits and vegetables have always been one of the most sought-after foods, which are rich in vitamins, minerals dietary fiber, and other nutrients. The most widely used fruit and vegetable-based food raw materials in food 3D printing are starch-based vegetables, including potatoes, taro, and yams. They are widely used in food 3D printing owing to their high starch content that confers them with suitable rheological properties.

Mashed potatoes are valuable fruit- and vegetable-based soft materials that can be used for the 3D printing of food. Liu et al.[38] directly used mashed potatoes for the 3D printing of food products and tested their rheological behavior. They found that mashed potatoes are pseudoplastic fluids with shear-thinning behavior, which is suitable for their application in 3D printing. They also explored the effect of potato starch addition on the printing characteristics of mashed potatoes. The addition of 2% potato starch yielded the best results to ensure the continuity of printing and to maintain a good resolution, as shown in Fig. 4A. In addition to the starch content that affects the printability of mashed potatoes, the processing conditions and formulation of mashed potatoes also have an impact on the printability of mashed potatoes. Martínez-Monzó et al.[39] explored the printability of mashed potatoes prepared with different ratios of milk and dehydrated mashed potatoes at 10, 20, and 30 °C. They found that the best printability can be achieved at 30 °C with 38 g of dehydrated mashed potatoes and 250 mL of whole milk.

In addition, some researchers investigated the effect of the addition of butter, alginate, and olive oil on the mechanical properties and rheology of mashed potatoes. They found that the addition of 1% butter to the microwave heated mashed potatoes can yield the best printability; the related printing results are shown in Fig. 4B[40]. This

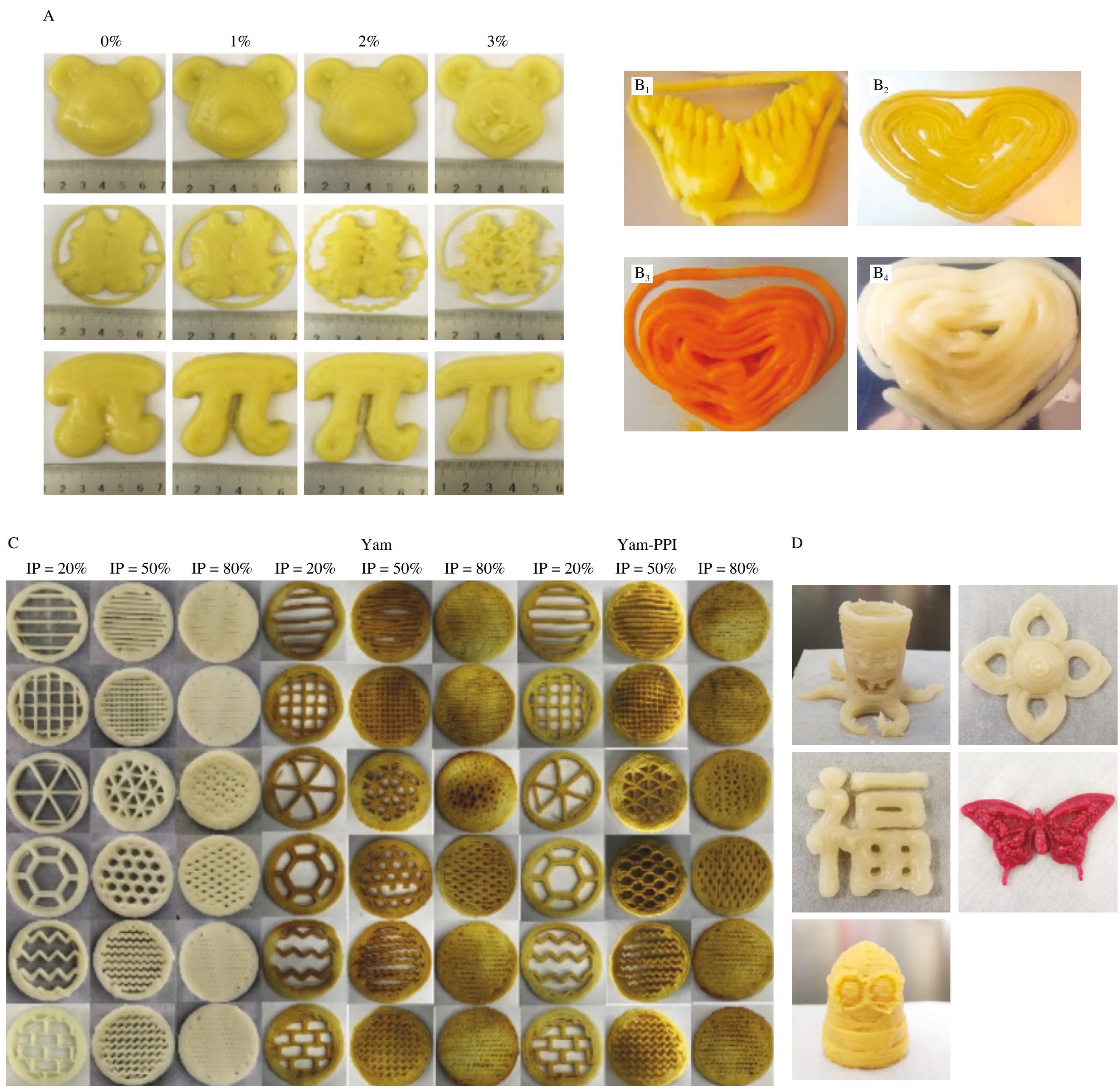


Fig. 4 (A) The effect of potato starch addition on the print quality of mashed potatoes (before frying)[38]. (B) Effect of the addition of agar, butter, and olive oil on the print quality of mashed potatoes after microwave heating[40]. B_1: 1% agar, B_2: 1% butter, B_3: 1/3 carrots, and B_4: boiled potato with 1% butter. (C) Printed yam snacks made from yam paste with different infill percentages (IP) levels (20%, 50% and 80%)[42]. (D) Related food products are printed from taro pulp[44].

can be attributed to the ability of butter to form lipid-starch complexes with starch molecules, which enhances structural stability and self-support. This is similar to the conclusion reached by Scheele et al.[41], who explored the effect of adding proteins and lipids on the texture, sensory properties, and printability of 3D-printed mashed potatoes and found that the addition of butter is the most attractive for both the printability and flavor enhancement of mashed potatoes.

In addition to mashed potatoes, starch-based viscoelastic materials such as yam paste and taro pulp can be used in food 3D printing. Feng et al.[42] used yam paste for printing yam snacks and explored the effect of the addition of potato processing byproducts on their printing characteristics. They found that all formulations had good printability, as shown in Fig. 4C. Wang et al.[43] also conduct 3D printing using yam paste as the raw material and obtained the best printability of yam paste with a moisture content of 35%. Huang et al.[44] examined the effects of additives and printing parameters on the printability of taro pulp using taro pulp as the printing material. They obtained the best printing results with the addition of 1% sodium alginate and printing parameters of 0.84 mm nozzle diameter, 25 mm/s nozzle movement speed, and 13 mm^3/s extrusion rate; the printing results are shown in Fig. 4D. Overall, starch-based raw fruit and vegetable materials are an integral part of our daily diet, and their printability should be further explored and optimized for future real-life production applications.

3.3 Other fruits and vegetables

Many raw fruit and vegetable materials used for food 3D printing, such as bananas, tomatoes, strawberries, guava, and carrots, have been used for preparing 3D-printed food[45-47]. These fruits and vegetables have higher nutritional value and more bioactive compounds than starch-based fruits and vegetables and are also more in line with people's understanding of a healthy diet. Therefore, the unique value

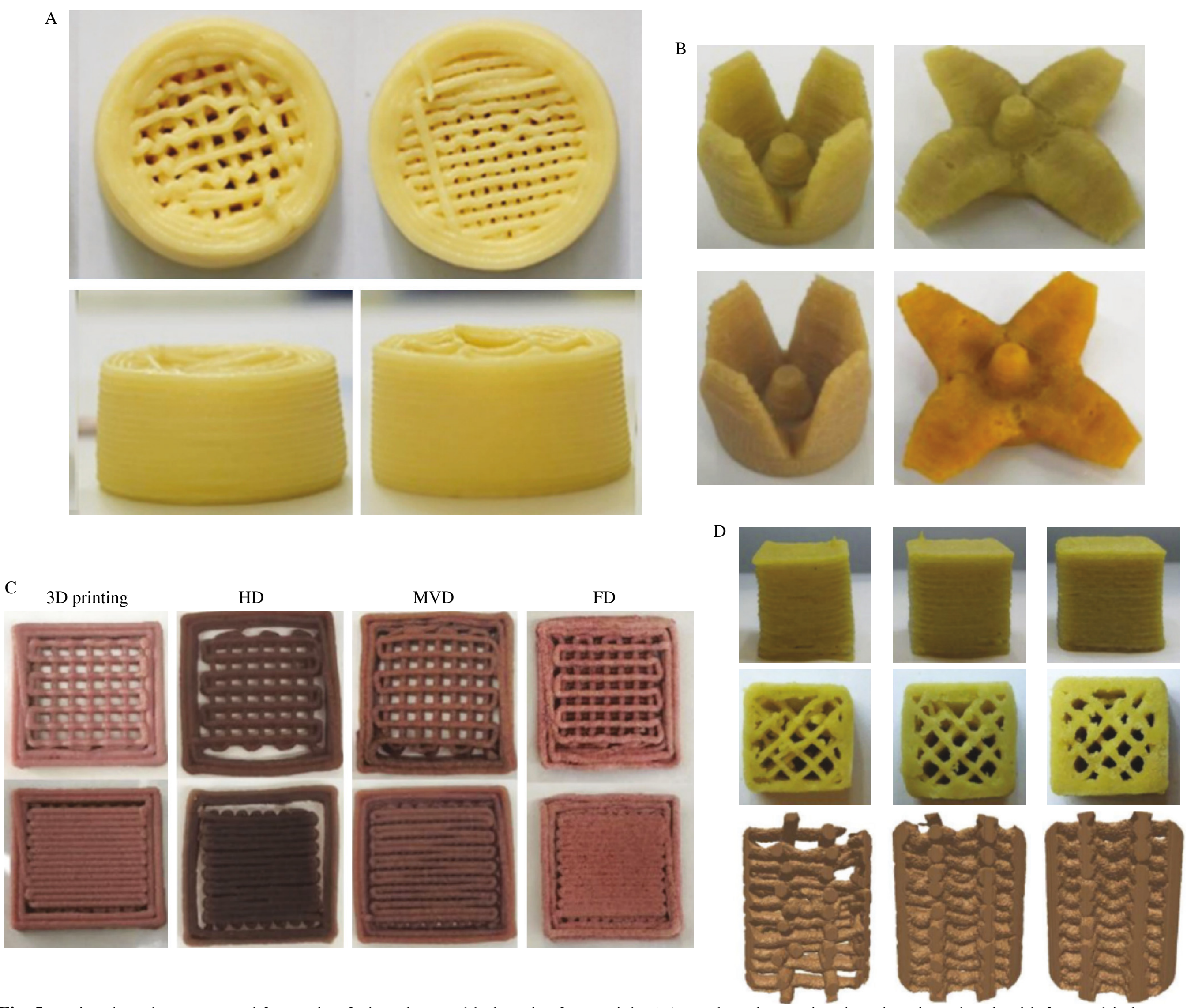


Fig. 5 Printed products prepared from other fruit and vegetable-based soft materials. (A) Food products printed on the wheat dough with freeze-dried mango powder[48]. (B) Food products printed from starch paste with added yellow peach powder[49]. (C) Fresh 3D printed sample and dried samples under different drying methods. HD: hot air drying; MVD: microwave vacuum drying; FD: freeze drying[50]. (D) 3D-printed functional food prepared from a mixture of fruit and vegetable raw materials[51].

of these raw materials should be explored to develop more delicious, healthy, and functional foods.

Because of the high moisture content of these foods, they cannot directly form viscoelastic materials. Therefore, they must undergo treatment or be used as auxiliary materials for food 3D printing[10], as shown in Fig. 5. Liu et al.[48] explore the effect of freeze-dried mango powder as an ingredient on the printability of wheat dough. They found the best printability for a flour: water: olive oil: freeze-dried mango powder ratio of 57.5:30:3:2.5 (*m*/*m*). The printing results are shown in Fig. 5A. Freeze-dried mango powder contains carbohydrates that increase the viscosity of the dough surface, allowing it to maintain a more stable structure. Guo et al.[49] conducted 3D printing by adding yellow peach powder to starch paste prepared from yam, purple potato, and buckwheat, and explored the effect of microwave treatment on the color, flavor, and deformation of the final product, as shown in Fig. 5B.

Feng et al.[50] used rose pollen as a printing material and mixed it with yam paste to prepare 3D printed food products with high polyphenols; the printing results are shown in Fig. 5C. They explored the effect of different post-processing methods on the quality of the products and the activity of bioactive compounds and found that products treated by microwave vacuum drying have the greatest advantage. Derossi et al.[51] developed different types and flavors of high nutritional value 3D printed foods by changing fruit and vegetable raw materials, which can provide 5%–10% of the daily energy, calcium, iron, and vitamin D requirements for children aged 3–10 years, as shown in Fig. 5D. These studies provide a well-informed channel for understanding the application of fruit and vegetable raw materials in functional 3D food production and offer a good reference to further explore the application of fruit- and vegetable-based raw materials in functional food 3D printing.

The above sections have explored the current status of the application of soft materials in food 3D printing. Although the printing parameters are the critical control points in the printing process of soft materials, due to the diversity of ink compositions, there is a wide range of parameter adjustments in the printing process, which will not be analyzed in detail here. Another extremely critical control point in the printing process of soft materials is the hygiene of the processing environment. The high moisture content of the soft material and the process of extrusion and shearing lead to an increase in ink temperature, which provides excellent conditions for microbial proliferation. Therefore, when printing soft materials, it is important to ensure environmental hygiene and clean printing equipment to avoid microbial growth, which could jeopardize food safety, as far as possible. Hygienic control of transport and storage also requires extra attention, and due to the textural characteristics of soft materials, transport, and storage should be as far as possible to avoid interference from external forces and maintain their structural characteristics.

4. Hydrogel materials

Hydrogels are a class of water-filled 3D polymer networks formed by the physical or chemical phase crosslinking of polymers[52]. They have received extensive academic and industrial attention in recent decades owing to their simple preparation methods, abundant raw materials, and tunable properties[53]. Benefiting from their wide range of raw materials, many plant-based raw materials, such as plant polysaccharides and plant proteins, have been used in hydrogel preparation and 3D printing[54-55]. This section focuses on the application of these plant-based food hydrogels in food 3D printing.

4.1 Starch-based hydrogels

Starch is a macromolecular polymer made of glucose linked by α-(1,4)-glycosidic and α-(1,6)-glycosidic bonds[56]. In addition, its molecular chains contain many hydroxyl groups that are susceptible to crosslinking reactions. When heated, the hydrogen bonds between the ordered and disordered starch molecules break and are dispersed in water. However, during cooling, the molecular chains are rearranged to form a starch gel network with large pore sizes through hydrogen bonding interactions. This makes them suitable for the preparation of hydrogel materials[57]. Potatoes, corn, wheat, and other starch-rich foods are widely used for preparing food hydrogels and as raw materials for 3D printing. The starch-based gels used for food printing are summarized in Table 1.

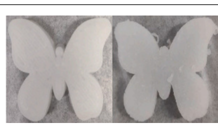
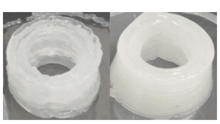
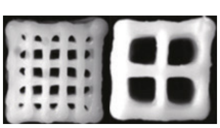
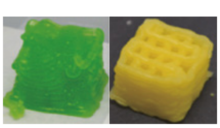
Zheng et al.[58] prepared starch-based hydrogels from wheat starch, potato starch, and corn starch and explored their applications in food 3D printing. The results showed that all three starch-based hydrogels can obtain complete 3D printed models, especially wheat starch, which presents good printability. However, single-starch-based hydrogels may suffer from lack of support, poor flowability, and poor resolution in practical applications. In response to these problems, many researchers have used different methods to optimize the printability of starch-based hydrogels.

Ma et al.[59] used radio frequency energy to treat potato starch gels with different moisture content to optimize the printability of the starch gels. They found that the radio frequency energy treatment successfully improves the printability of potato starch gels, and the improvement effect is especially obvious on starch gels with 70% moisture content. This can be attributed to the depolymerization of the long chains of straight-chain starch into short chains with radio frequency energy treatment. The reduction in molecular weight reduces the viscosity of the starch gel, making it more suitable for 3D printing. Maniglia et al.[60] conducted pulsed electric field (PEF) treatment of wheat starch and tapioca starch hydrogels and found that PEF treatment improves the printability of wheat starch hydrogels. The mechanism of action is similar to that of radio frequency energy treatment of potato starch, which is achieved through the depolymerization of starch granules. In addition to the above two treatments, studies have shown that ozone processing, dry heat treatment, microwave treatment, and ultrasound have positive effects on the printability of starch-based gels[61-63].

These methods are primarily used to optimize the printability of starch-based hydrogels through physical modifications. Many researchers have used additives to improve the printability of starch-based hydrogels; polysaccharides are common additives. Cui et al.[64] improved the printability of potato starch gels using sodium alginate and xanthan gum. They found that the composite hydrogel containing potato starch, sodium alginate, xanthan gum, and water at a mass ratio of 6:2.5:2.5:89 exhibited the best printability. Xiao et al.[65] and Yuris et al.[66] explored the effects of *Mesona chinensis* polysaccharides on starch gel properties. The results of these studies indicate that the addition of *M. chinensis* significantly affects the rheology, strength, and hardness of the starch gels, which are important indicators for determining the printability of the gels. Furthermore, chitosan, guar gum, carrageenan gum, and Arabic gum have been used to optimize the printability of starch-based hydrogels[67-68]. Therefore, we can draw on the related studies to design the starch-based hydrogel inks required for their applications, thus further expanding the application space of starch-based hydrogels in food 3D printing.

In addition to polysaccharides, proteins, and salts can also be used to improve the printability of starch-based hydrogels. The addition of proteins to starch-based hydrogels is mainly because of their unique gelation properties, which can improve the stability and resolution of the printed structure of starch-based hydrogels[63]. Ji et al.[69] improved the printability of tapioca starch using casein and found that the best printability of gel samples was achieved when the amount of casein added was 10% of the starch mass. This is because a high concentration of casein reduces the viscosity of the hydrogel system and increases its yield stress, facilitating smooth extrusion and mechanical strength of the gel. It is beneficial to further understand the interactions between proteins and starch to develop new composite starch-based hydrogel materials. Salts can also be used to modify starch-based hydrogels and improve their printability by affecting the pasting and regenerative properties of starch[61,70].

Table 1
Starch-based hydrogels for food 3D printing.

Materials	Products	Print parameters	References
Wheat starch hydrogels		Nozzle height: 1.00 mm Nozzle diameter: 1.20 mm Print speed: 30.00 mm/s	[71]
Potato starch hydrogels		Nozzle height: 1.00 mm Nozzle diameter: 1.00 mm Print speed: 5.00 mm/s	[72]
Buckwheat starch-pectin hydrogels		Nozzle height: unannounced Nozzle diameter: 0.80 mm Print speed: 20.00 mm/s	[61]
Corn starch hydrogels		Nozzle height: unannounced Nozzle diameter: 0.80 mm Print speed: 0.30 mm/s	[73]
Cassava starch hydrogels		Nozzle height: 0.80 mm Nozzle diameter: 1.00 mm Print speed: 8.00 mm/min	[74]
Rice starch-catechin hydrogels		Nozzle height: 1.00 mm Nozzle diameter: 0.80 mm Print speed: 50.00 mm/s	[75]
Anthocyanin-potato starch hydrogels		Nozzle height: 0.85 mm Nozzle diameter: 0.85 mm Print speed: 25.00 mm/s	[76]

Guo et al.[61] studied the effect of Ca^{2+} on the printing performance of a buckwheat starch-pectin system. They found that the printing accuracy of the microwave-treated buckwheat starch-pectin system was significantly improved when 1% mass fraction of calcium chloride was added. This is because the introduction of Ca^{2+} causes the buckwheat starch-pectin system to agglomerate, reducing the viscosity of the composite hydrogel and making it easier to extrude, thereby improving its printing accuracy. Zheng et al.[70] investigated the effect of NaCl on the printing quality of wheat starch gels from the viewpoint of starch pasting and regeneration. They showed that wheat starch gels have the best printing quality when 150 mmol/L NaCl solution is added because the NaCl solution at this concentration can regulate the viscoelasticity of starch gels to a moderate value by affecting the pasting temperature of starch such that they can have good extrudability and maintain structural stability after extrusion.

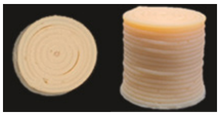
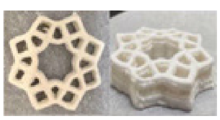
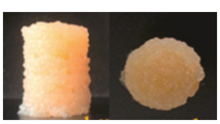
4.2 Protein-based hydrogels

Protein is a common biological resource in nature and an essential nutrient in food. Hydrogel systems constructed using plant proteins show higher sustainability and lower cost and are also capable of structural and functional modulation[77]. This makes them promising for a wide range of applications in the field of food 3D printing. However, the stability and mechanical strength of pure protein hydrogels are poor[78], making them unsuitable for food 3D printing. Therefore, at present, mostly composite hydrogels of proteins and polysaccharides are used for food 3D printing. Some of the protein-based gels 3D printed food are shown in Table 2.

Yu et al.[79] formulated emulsion gels from soy protein isolate (SPI) to investigate the effects of different concentrations and polysaccharide types on the printability of the SPI emulsion gels. The results verified that the addition of guar gum and xanthan gum improved the gel printing performance of the SPI emulsion. In addition, the printability, viscosity, and gel strength of the composite hydrogel system increased with increasing polysaccharide concentration. This may be because the introduced polysaccharides can crosslink with protein molecules through hydrogen bonding, which increases the mechanical strength of the composite gel. Some scholars have improved the printability of protein-based hydrogel systems using sodium alginate and carrageenan[80-81]. Although their addition has improved the printability of protein gels, some studies have shown that the addition of such hydrocolloids can have adverse effects on human health[82].

Therefore, functional polysaccharides isolated from foods have been used to improve the printability of protein gels and impart certain functionalities to foods. Pan et al.[55] improved the printability of soy protein hydrogels by introducing *Flammulina velutipes* (FV) polysaccharides. They showed that the addition of FV polysaccharides improved the printability of soybean protein gel by changing the secondary structure of the protein. Xu et al.[83] studied the function of protein gel-printed foods by adding modified apricot polysaccharides to SPI gels. The results indicated that apricot polysaccharides not only improved printability but also conferred lipid-lowering ability. Some researchers have encapsulated probiotics using emulsion gels prepared from tea protein and xanthan gum and have successfully obtained 3D-printed foods with good probiotic activity[84].

Table 2
Protein-based hydrogels for food 3D printing.

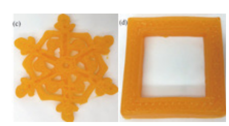
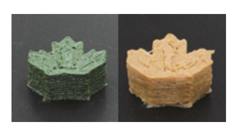
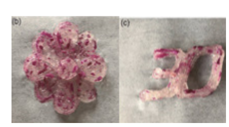
Materials	Products	Print parameters	References
Pea protein hydrogels		Nozzle height: 1.00 mm Nozzle diameter: 1.10 mm Print speed: 20.00 mm/s	[88]
Tea protein-xanthan gum hydrogels		Nozzle height: unannounced Nozzle diameter: 0.84 mm Print speed: 25.00 mm/s	[84]
Peanut protein-purple sweet potato flour hydrogels		Nozzle height: unannounced Nozzle diameter: 0.40 mm Print speed: 20.00 mm/s	[89]
SPI-xanthan gum hydrogels		Nozzle height: unannounced Nozzle diameter: 1.20 mm Print speed: 15.00 mm/s	[90]
Peanut protein isolate-vegetable powder hydrogels		Nozzle height: 0.80 mm Nozzle diameter: 0.80 mm Print speed: 20.00 mm/s	[91]
Faba bean protein isolate emulsion hydrogels		Nozzle height: 0.50 mm Nozzle diameter: 0.84 mm Print speed: 25.00 mm/s	[92]
FV polysaccharide-soy protein hydrogels		Nozzle height: 1.00 mm Nozzle diameter: 1.20 mm Print speed: 15.00 mm/s	[55]

The integration of protein gel systems and 3D printing has also been extensively implemented in the development of plant-based meat. This innovative approach has allowed for the generation of plant-based meat alternatives that closely resemble the microfibrillar and anisotropic structure of animal meat, thereby providing consumers with a highly appealing and sustainable dietary option. In the study conducted by Ko et al.[85], they achieved successful 3D printing of beef analogs using a gel system incorporating SPI and various polysaccharides, employing a coaxial nozzle-assisted printing method. The printed meat alternatives exhibited a texture that closely resembled that of actual beef when the ink formulation contained carrageenan and glucomannan at concentrations of 2.5% and 1.5% (*m*/*m*), respectively. In a similar vein, Chen et al.[86] conducted research on the development of simulated meat products using a sophisticated gel system incorporating soy protein and polysaccharides. These printed products were subsequently subjected to deep-frying. The findings of their study disclosed that the geometric pattern and filling ratio played a crucial role in determining the texture of the fried samples. Specifically, the prints with a triangular filling pattern and a 60% filling ratio closely resembled the texture of shredded deep-fried chicken breasts. This highlights the significance of optimizing both the shape and the amount of filling in achieving desired textural attributes in plant-based meat analogs. In addition to texture simulation, 3D printing can give plant meat a variety of shapes and customized nutritional components that are not available in traditional meat products, making it more suitable for consumer demand and having a broad scope for future development. In addition, the protein gel system has a wide range of applications in the fabrication of cell culture scaffolds. Su et al.[87] successfully prepared protein fiber scaffolds similar to pig muscle tissues by using the grain alcohol soluble protein gel system as an ink in combination with high-precision 3D printing technology, and ultimately succeeded in cultivating slices of artificial pork on the scaffolds.

4.3 Other plant-based hydrogels

In addition to the above two hydrogel materials, some fruits and vegetables are also present that cannot be prepared as hydrogels alone owing to low protein and starch content and high moisture content[93]. These materials require some processing before they can be prepared as hydrogels for 3D printing; the relevant printouts are shown in Table 3.

Table 3
Other plant-based hydrogels for food 3D printing.

Materials	Products	Print parameters	References
Mashed potatoes/ strawberry juice hydrogels		Nozzle height: unannounced Nozzle height: 1.20 mm Print speed: 25.00 mm/s	[99]
Garden pea and carrot hydrogels		Nozzle height: 0.50 mm Nozzle diameter: 0.84 mm Print speed: 25.00 mm/s	[100]
Orange concentrate-starch hydrogels		Nozzle height: unannounced Nozzle diameter: 0.83 mm Print speed: unannounced	[101]
Spinach and carrot hydrogels		Nozzle height: 0.90 mm Nozzle diameter: 1.00 mm Print speed: 25.00 mm/s	[95]
Rose petal hydrogels		Nozzle height: 0.80 mm Nozzle diameter: 1.50 mm Print speed: 25.00 mm/s	[97]

Lipton et al.[94] prepared a celery juice hydrogel using agar and successfully used it in food 3D printing. However, owing to the high moisture content of celery juice, a long gelation time is required after printing before the printed structure has some strength. To address this issue, researchers often use freeze-dried fruit and vegetable powders to prepare hydrogels to reduce the water content and enhance the stability of printed structures. Compared with fruit and vegetable concentrates, freeze-dried fruit and vegetable powders are more widely used in the preparation of fruit- and vegetable-based hydrogels, and their lower moisture content makes them an ideal choice for storage and transportation. Kim et al.[95] prepared fruit- and vegetable-based hydrogels by adding different hydrocolloids to solutions prepared from freeze-dried powders of broccoli, spinach leaves, and carrots, and explored the effect of different hydrocolloids on the print quality. The printing results showed that the prints maintained excellent extrudability and high resolution even when the mass fraction of freeze-dried vegetable powder in the hydrogel reached 30%. The higher solid content in the prints facilitated the post-print structure retention and storage period[96].

Feng et al.[97] used dried rose petal fragments and sodium alginate to prepare the gel material. They investigated the effect of size of the rose petal fragments and the addition amount on the printing quality. The results showed that when the petal fragments were less than 0.5 cm in diameter and the addition amount was less than 6%, the product could be prepared with exquisite shape and strong rose flavor. This review provides a reference for the future printing of heterogeneous raw materials and further expands the range of raw materials for preparing fruit- and vegetable-based hydrogel materials.

Although fruits and vegetables with high moisture content are not suitable for direct preparation into gels for the 3D printing of conventional foods, they are suitable for the preparation of dysphagic patient foods, benefiting from the fluffy structure and lower hardness of their prints[98]. Therefore, when we adopt 3D printing technology for food production, we must consider the needs of the target population to flexibly adjust indicators such as ink rheology and the mechanical properties of prints.

The printing process parameters and food safety control methods for hydrogel materials are similar to soft materials. However, what he should consider is the customer acceptance, although in our dietary system has long been involved in the gel food, such as casein gel system and collagen gel system, but people usually still associate gel with unhealthy. How to get the public to accept gel-based food is the most important thing to promote the development of food 3D printing, and it is also the direction that we need to pay extra attention to while developing the technology. After all, customer acceptance is the ultimate goal and core driver of food development.

5. Prospects and challenges

Combining plant-based food raw materials with 3D printing technology is an important direction for the future of food manufacturing, which offers the benefits of personalized customization and nutritional health. This article classifies the application of various substances in food 3D printing based on their different states and rheological properties and establishes a clearer connection between print quality and the properties of the printing inks. Although plant-based raw materials, such as starch, functional polysaccharides, polyphenols, and plant-based proteins, the print quality of current ink systems is often insufficient to support commercialization, with delamination marks and inadequate structural support and integrity still common issues. Moreover, the diversity of plant-based materials and their properties can result in significant differences in print quality, even when the composition ratio remains constant. This makes it difficult to unify and standardize printing parameters, which hampers the promotion of food 3D printing technology for household and personalized use. Further development of safety studies related to food 3D prints is also necessary. The printing process and ink pretreatment may involve additional steps do not present in traditional food processing, particularly when modifying raw material components. However, current research on 3D printing of food products primarily focuses on print preparation, lacking comprehensive safety evaluations. Therefore, it is crucial to enhance the development of plant-based inks, optimize their printing performance, ensure the safety of the modified inks, and establish a comprehensive database relating to different inks alongside their corresponding optimal printing parameters. By promoting the use of plant-based food materials in the realm of food 3D printing, we can accelerate the commercialization of 3D printed food and create future food products that embody a fusion of flavor, nutrition, and environmentally sustainable advantages.

Declaration of competing interest

Authors declare that they have no conflict of interest.

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Authors contribution

Zhihao Liu: Conceptualization, investigation, writing-original draft. Xinna Hu: Conceptualization, investigation. Shuyu Lu: Investigation. Bo Xu: Investigation. Chenyu Bai: Formal analysis. Tao Ma: Conceptualization, formal analysis, writing-reviewing and editing. Yi Song: Supervision, funding acquisition.

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