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REVIEW



Novel evaluation technology for the demand characteristics of 3D food printing materials: a review

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ABSTRACT

As a recently developed way of food manufacturing - 3D printing - is bringing about a revolution in the food industry. Rheological and mechanical properties of food material being printed are the determinants of their printability. Therefore, it is important to analyze the requirements of different 3D printing technologies on material properties and to evaluate the performance of the printed materials. In this review, the printing characteristics and classification of food materials are discussed. The four commonly used 3D printing techniques e.g. extrusion-based printing, selective sintering printing (SLS), binder jetting, and inkjet printing, are outlined along with suitable material characteristics required for each printing technique. Finally, recent technologies for evaluation of 3D printed products including low field nuclear magnetic resonance (LF-NMR), computer numerical simulation, applied reference material, morphological identification, and some novel instrumental analysis techniques are highlighted.

KEYWORDS

3D food printing; 3D printed types; material characteristics; rheology; quality of 3D printed products; simulation models

Introduction

Three-dimensional (3D) printing, known as additive manufacturing (AM), solid freeform fabrication (SFF), is a construction method layer by layer in three - dimensional space (Fahmy, Becker, and Jekle 2020; Lamichhane et al. 2020). Under the control of a computer, the product can be accurately and rapidly molded using a 3D digital model developed in advance. Since its emergence, 3D printing has been regarded as a potential industrial revolution, and has been gradually applied in construction, manufacturing, healthcare, education and aerospace (Liu et al. 2018). In the food sector, 3D printing technology has been used for the first time by researchers at Cornell University to make food. They developed the simple and inexpensive Fab@Home project to promote the use of 3D printing technology for individuals, allowing people to print out foods such as chocolate at home (Malone and Lipson 2007). Figure 1 shows the Fab@Home Model 1 developed by Cornell University. 3D food printing has a number of advantages, including customizing food, producing more complex food shapes, accurate nutrition ratios, simplifying supply chains, reducing waste and expanding the range of ingredients (Liu et al. 2017; Piyush and Kumar 2020). For instance, 3D printing can help elderly people with chewing and swallowing difficulties customize food (Dick, Bhandari, and Prakash 2019a), and it is also possible to increase children's interest in food by printing a variety of interesting and complex shapes of

food, and to provide children with a balanced nutrition by adjusting the formula (Derossi et al. 2018). In recent years, many food materials have been reported for 3D printing, including chocolate (Mantihal et al. 2019; Mantihal, Prakash, and Bhandari 2019), meat (Dick et al. 2020; Dick, Bhandari, and Prakash 2019a), cheese (Le Tohic et al. 2018), milk protein (Liu et al. 2018), fruits and vegetables (Derossi et al. 2018; Severini et al. 2018), cereal-based product (Fahmy, Becker, and Jekle 2020), and mashed potatoes (Feng et al. 2020; Liu et al. 2020), some of which have been applied in actual industrial production.

3D printing technology involves a variety of technical principles. Currently, techniques applied to the food sector generally include extrusion printing, selective sintering printing (SLS), ink-jet printing and binder jet printing (Baiano 2020; Liu et al. 2017). Among them, the most commonly used is extrusion printing, because it is suitable for the rheological properties of more food materials (Dick, Bhandari, and Prakash 2019a). No matter what kind of printing technique is adopted, it has strict requirements on the characteristics of food to meet the printing needs. 3D printing was originally used in the manufacture of non-food products, including metals, ceramics, polymers and so on (Zhou, Fu, and He 2020). Since there is no need to consider food safety when printing these materials, extreme temperature conditions can be applied, some organic solvents, organic crosslinking agents and other additives that do not meet food safety standards can be used. Nevertheless, when 3D



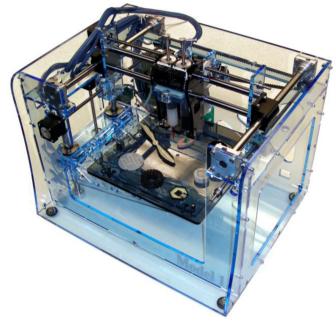


Figure 1. Fab@Home Model 1 for 3D food printing, one of the first generation of 3D food printer, available at https://www.fabathome.net/.

printing is used for food, the application of these conditions is limited, which requires higher quality of printing materials themselves. Moreover, due to the complex composition of food raw materials and the differences in physical and chemical properties of each component, 3D printing of food its own characteristics in material selection (Gholamipour-Shirazi et al. 2020), which also brings challenges to the application of 3D printing technology in food processing. Therefore, one of the difficult problems of 3D printing food is how to choose and optimize food materials, so that they can better combine with printing technology. In general, materials used for 3D food printing need to be extrudable and supportable. Good extrudability can ensure the smooth extrusion of food materials from the nozzle, and good supposition can ensure that each layer of printing does not deform or collapse (Yang et al. 2019). Godoi et al. proposed three key factors related to the properties of food materials, namely printability, applicability and post-processing, which provided a basis for the optimization of food materials in 3D printing (Godoi, Prakash, and Bhandari 2016). Prakash et al. showed that it is important to understand the properties and composition of food materials in 3D food printing. They proposed two main bases for 3D food printing, one is the fluidity of food materials when squeezed through a nozzle, and the other is the ability of food materials to maintain shape during and after printing (Prakash et al. 2019). Liu et al. also showed that it is necessary to fully understand the material properties before building three-dimensional structures. Moreover, they proposed four factors affecting the printing accuracy, one of which is the material properties, such as rheological property, gelation, melting and glass transition temperature (Tg) (Liu et al. 2017). Dankar et al. found that evaluating the properties of food materials, especially the rheological properties, is very important for predicting 3D printing design and flow conditions, such as pump and syringe sizes, printing time and other parameters (Dankar et al. 2018).

Obviously, evaluating and understanding the characteristics of food material such as rheological, physicochemical, structural, and mechanical properties is essential for accurate food printing. There are many existing techniques for detecting food printing characteristics, such as rheometer, Texture profile analysis (TPA), Scanning electron microscopy (SEM), Differential scanning calorimeter (DSC), Fourier transform infrared (FT-IR), wide angle X-ray diffraction (XRD), etc (Liu et al. 2020; Wang et al. 2018; Yang et al. 2018). However, these techniques are not enough to fully meet the detection requirements well. The development of 3D food printing technology always involves the updating of material feature detection and characterization methods. In recent years, a number of studies have focused on the introduction of new techniques for the updating of detection methods. It is worth noting that the components of each kind of food will be different, and the 3D food printing process will behave differently, which makes the detection work very complicated and passive. Therefore, some researches focus on the use of computer simulation methods to predict the printing characteristics of food materials, such as numerical simulation methods (Liu et al. 2020), constitutive models (Jonkers, van Dommelen, and Geers 2020) and so on. The application of these new techniques has further developed the detection technology of 3D food printing. It is valuable to collect, collate and analyze these evaluation techniques in 3D food printing.

Actually, there have been several reviews involving the topic of 3D food printing from different perspectives. Voon et al. reviewed the inks suitable for 3D food printing and their development status (Voon et al. 2019). The review published by Dankar et al. focused on the benefits and limitations of 3D food printing, and how to optimize the printing parameters for different food ingredients (Dankar et al. 2018). The review presented by Godoi et al. mainly introduced various printing techniques, as well as the basic ingredients of food and the feasibility of 3D printing (Godoi, Prakash, and Bhandari 2016). The review presented by Liu et al. analyzed several key factors for achieving accurate printing and reviewed the applications of 3D printing in military and space food, elderly food, and confectionery market (Liu et al. 2017). Another review published by Sun et al. mainly introduced various food printing platforms, printable materials and recipes, various printing techniques (Sun et al. 2015). However, there is little knowledge about the properties of 3D printed materials and new evaluation techniques. In the present review, the printing properties of food materials required by different printing techniques are analyzed, and some new techniques for evaluating the properties of 3D food printing materials are highlighted, which can provide reference for the ongoing evaluation techniques in the research field of 3D food printing.

Printability of different food materials

Printability is a feature that refers to how the characteristics of a material adapt to printing processing and deposition

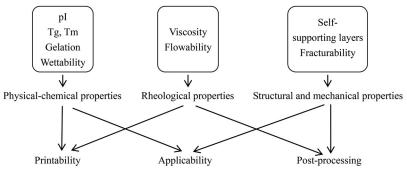


Figure 2. Material characteristics to be considered in 3D food printing.

and maintain its structure after printing. These characteristics include viscosity and rheological properties of molten solid and liquid matrix materials, gelatability and thermal properties of materials used in extrusion printing technology, as well as wettability, particle size distribution, fluidity and bulk density of powder matrix materials. In addition, printed food usually needs to undergo subsequent processing, so materials used for 3D printing also need good postprocessing performance, such as resistance to cooking, baking and frying (Dankar et al. 2018; Jiang et al. 2019). Figure 2 shows the material characteristics that need to be considered to improve the printability of food materials in 3D printing.

Although 3D food printing technology continues to improve, and food material properties can be modified in more ways, the materials available for 3D printing are still limited. Sun et al. classified printable food materials into three categories based on their printable characteristics, namely native printable food materials, non-native printable traditional food materials, and alternative ingredients. Native printable food materials refer to those materials that have good rheological properties. They can be extruded smoothly from the extrusion nozzle and can maintain a certain shape after deposition (Cohen et al. 2009). Non-native printable traditional food materials often lack these characteristics and cannot be printed directly (Saunders 2020). Alternative ingredients usually refer to those that have good nutrients but are not easy to consume directly as food, such as proteins in insects and residues from food processing. In 3D food printing, these alternative materials are mixed with some traditional ingredients to serve as the printing substrate (Izdebska and Zołek-Tryznowska 2016). Table 1 shows the categories of food printability. In the category of native printable food materials, the ease of printing varies according to the properties of the materials. Kim et al. selected methyl cellulose with different concentrations to simulate the extruded 3D printing characteristics of food, and classified the printed food by evaluating the deformation behavior and processing characteristics of the sample. The extrusion properties of the sample can be classified into four types: easy, normal, difficult and extremely difficult. The corresponding printing characteristics are suitable for complex 3D printing, suitable for ordinary 3D printing, extruders requiring high output, and melting (Kim, Bae, and Park 2017).

However, people's daily staple foods, such as fruits and vegetables, rice, meat, do not have the characteristics of direct printing, they belong to the non-native printable traditional food materials. These materials can be printed by adding some hydrophilic colloid to improve their rheological and mechanical properties (Park, Kim, and Park 2020). There have been numerous reports of research in this area. Liu et al. took milk protein as the printing material and added different concentrations of whey isolated protein (WPI) to it to evaluate the effect of WPI on the printing performance of milk protein. The results showed that WPI could reduce the apparent viscosity of milk protein, soften the milk protein paste, and facilitate the printing process (Liu et al. 2018). By adding sodium alginate and gelatin to soy protein isolate (SPI), Chen et al. developed a hybrid food substrate as a material for 3D food printing. The addition of gelatin and sodium alginate enhances the printing performance of SPI, improves the hardness and chewability of the product (Chen et al. 2019). Liu et al. also found that rice paste mixed with a certain concentration of sodium alginate could be an ideal 3D printing material (Liu et al. 2020). Wang et al. showed that the addition of NaCl could improve the rheological properties of surimi gel and enhance its printability (Wang et al. 2018). Liu et al. found that mashed potatoes showed good extrudability and printability after adding 2% potato starch (Liu et al. 2018). Feng et al. found that a certain amount of potato processing byproducts can improve the 3D printing characteristics of yam (Feng et al. 2020). In addition to adding additives, the printing performance of food materials can also be improved by adjusting the food formula. Pulatsu et al. investigated the printability and post-processing capacity of cookie dough by adjusting the recipes for fat, flour, milk and sugar. The results showed that the best sample composition was 37.5 g sugar, 62.5 g fat, 100 g tapioca flour, and 32.5 g milk, which were easy to print, stable in shape and good in appearance, and would not collapse after baking (Pulatsu et al. 2020). Yang et al. studied the effects of different ingredients on the physical properties of 3D printed dough. The results showed that the gel properties and physical properties of the dough were optimal when the formula contained 29 g of water, 6.6 g of sucrose, 6.0 g of butter, 48 g of flour and 10.4 g of egg per 100 g (Yang et al. 2018). It can be seen that nonnative printable traditional food materials can be printed by selecting appropriate additives. Nevertheless, it is a tedious and challenging job to adjust one by one according to the

Table 1. Categories of food printability and corresponding material characteristics.

Categories	Characteristics	Representative food	Printability	Ref.
Native printable	Materials with good rheological	Chocolate	Easy	(Mantihal et al. 2017)
food materials	properties and mechanical	Cheese	Easy	(Le Tohic et al. 2018)
	properties. They can be	Bean paste	Normal	(Kim, Bae, and Park 2017)
	printed directly without	Dough	Difficult	(Liu et al. 2019)
	additional additives.	Sugar paste	Difficult	(Kim, Bae, and Park 2017)
Non-native printable traditional food materials	Traditional staple foods lacking direct printing characteristics.	Meat	Need additives	(Dick, Bhandari, and Prakash 2019b)
	Their printing requires additional additives such as hydrophilic colloid to regulate	Rice		(Anukiruthika, Moses, and Anandharamakrishnan 2020)
	the rheological properties of	Fish		(Wang et al. 2018)
	the extrusion process and the mechanical behavior of the deposition process.	Fruits and vegetables		(Severini et al. 2018)
Alternative ingredients	Potential nutrients that can replace traditional food sources. They are new sources	Edible insect powder Derivatives from algae, seaweed, fungi	Alternative	(Severini and Derossi 2016) (Sun et al. 2015)
	of functional ingredients in food, such as proteins and fibers isolated from insects, algae, etc. These alternative ingredients can be made into 3D-printed pastes or powder that can be used in the production of custom meals.	Residues from agricultural and food processing		(Chinga-Carrasco et al. 2019)

characteristics of different foods. Cohen et al. developed a hydrocolloidal printing to customize food 3D printing. This process uses a mixture of xanthium gum and gelatin as the base material, and then mixes different flavor agents to achieve the independent adjustment of texture and flavor. With less use of this mixed material, a variety of foods can be simulated and printed (Cohen et al. 2009). This opens up a new way of choosing materials for 3D food printing.

Characteristics of food materials for different 3D printing techniques

There are different 3D printing techniques can be used in food. Each technique has its own scope of application. It is necessary to choose the appropriate printing technique according to the characteristics of food materials to meet the needs of accurate printing. In addition, some methods should be used to change the composition of food materials to suit specific printing techniques, such as adding some additives under the premise of ensuring food safety. In industry, 3D printing technology can be divided into seven categories, including material extrusion, material jetting, binder jetting, powder bed fusion, vat photopolymerization, directed energy deposition, and sheet lamination (Zocca et al. 2020). However, due to the diversity of physical and chemical properties of food materials, not all of these techniques are suitable for 3D printing of food. As mentioned in the introduction section above, 3D printing technique that can be used in food mainly includes extrusion based printing, selective sintering printing (SLS), binder jetting, and inkjet printing (Hao et al. 2010; Huang et al. 2013). Table 2 lists 3D food printing techniques and characteristics of food materials required for printing. Each of these techniques for 3D food printing and the characteristics of food materials required are discussed in detail in subsequent sections.

Extrusion based printing and characteristics of food materials required

Compared with other techniques, extrusion based 3D printing is a relatively simple process that has been widely used. The main component of extrusion based 3D printing is a nozzle with a piston, which has food ink inside. The ink is extruded from the nozzle by the piston, and accumulates layer by layer on the printer bed. In the printing process, the moving path of the nozzle is controlled by a preset 3D model, so that the extruded printing material can produce a specific shape after solidification (Sun et al. 2015). Figure 3 shows a kind of schematic diagram of extrusion based printing. Depending on the extrusion temperature setting, this printing technique can be classified as room temperature extrusion (RTE), hot-melt extrusion (HME) and hydrogelforming extrusion (HFE) (Sun et al. 2018), which are suitable for a variety of food materials that exhibit different aggregation states at room temperature. RTE is suitable for food materials that have fluidity at room temperature and can be squeezed out of a nozzle, such as dough, starch paste, cellulose paste, cheese, etc (Álvarez-Castillo et al. 2021; Feng et al. 2018; Kern, Weiss, and Hinrichs 2018; Lille et al. 2018; Thibaut et al. 2019). Also known as fused deposition modeling (FDM), HME was first introduced by Crump in 1991 (Crump 1991; Sun et al. 2015). In contrast to the RTE, the HME contains a heating element that allows the ink to be heated as it passes through the nozzle. According to the different characteristics of material, the appropriate heating temperature is set to maintain the ink melting state and make it through the nozzle smoothly. Because the heat tends to be slightly higher than the material's melting point, the extruded ink can quickly solidify to produce the desired 3D -printed shape, which is also an advantage of HME technique (Mantihal, Kobun, and Lee 2020). HME is widely used in various 3D printing fields, both solid and semi-solid

Table 2. List of 3D food printing techniques and characteristics of food materials required for printing.

Technique		Principle	Fabricated materials	Material characteristics	Ref.
Extrusion based printing	Hot-melt extrusion	Extrusion and deposition	Chocolate, confection	Solid and semi-solid that can be melted by heating, solidify immediately after extrusion, with good rheological and mechanical properties	(Duty et al. 2018; Lanaro, Desselle, and Woodruff 2019; Long et al. 2018)
	Room temperature extrusion	Extrusion and deposition	Dough, starch paste, cellulose paste, cheese, hummus, jelly	Natively printable, have fluidity at room temperature, with certain mechanical properties and can be formed stably after deposition	(Feng et al. 2018; Lille et al. 2018; Yang et al. 2018)
	Hydrogel- forming extrusion	Extrusion and deposition	Xanthan gum and gelatin	Appropriate polymer rheological properties and gel forming mechanism	(Cohen et al. 2009; Serizawa et al. 2014; Sun et al. 2018)
Selective sintering printing	Selective laser sintering (SLS) Selective hot air sintering and melting (SHASAM)	Powder binding and heat source (laser) Powder binding and heat source (hot air)	Sugar, fat based materials	Low melting powder, suitable particle compressibility, particle density and particle wettability, free flowing	(Berretta et al. 2013; DePalma et al. 2020; Liu et al. 2020)
Binder jetting	sang (amaun)	Bonding powder through liquid binder	Sugar powder, flour, starch	Powder, appropriate shape, appearance, size, distribution, fluidity, and accumulation density of powder particles	(Mostafaei et al. 2020; Wegrzyn, Golding, and Archer 2012)
Inkjet printing	Continuous inkjet printing (CIJ) Drop on demand printing (DoD)	Liquid extrusion and dispersive deposition Drop-on-demand deposition	Butter, cream, chocolate, jam, as decorations	Low viscosity, appropriate physical properties such as surface tension and inertia of liquid materials	(Daly et al. 2015; Murphy and Atala 2014)

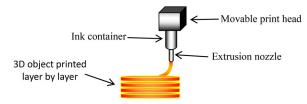


Figure 3. Schematic diagram of extrusion based printing.

can act as its ink (Sun et al. 2018). In the early stage, HME is mainly used in the production of plastic and rubber products (Chokshi and Zia 2004). These materials are easy to melt when heated and solidify when cooled, which is suitable for hot extrusion printing. In the food field, HME has been paid more and more attention, it has successfully produced personalized 3D chocolate products (Hao et al. 2010; Karavasili et al. 2020; Zoran and Coelho 2011). By controlling the temperature of food printing head, Bocusini achieves the purpose of using a printer to print five kinds of food, including confectionary, meat, snacks, fruits and vegetables, and dairy products. The operator can simply replace the printed capsules or assemble them on demand to complete the vision of printing a variety of delicious food at home (O'Neal 2015). HFE is a novel 3D printing method for food. It uses a specific device to squeeze hydrocolloid solution or dispersion into a polymer/hardening/gel setting bath. In the HFE process, appropriate polymer rheological properties and gel forming mechanism, as well as solution temperature control are important, which are the key to

successful printing (Sun et al. 2018). HFE technique has been used for commercial food production. For example, it uses fruit puree and other gel as materials to print soft food, which is suitable for elderly people with swallowing problems (Le-Bail, Maniglia, and Le-Bail 2020; Serizawa et al. 2014).

The characteristic of food material is the key factor to choose which technique to adopt. Le-Bail et al. proposed three parameters should be considered for the extrusion technique: (1) Food ink should have appropriate rheological properties so that it can be successfully extruded from the nozzle. (2) The extruded food ink should have good mechanical properties to support the 3D structure of the printed layers and keep them from collapsing. (3) After deposition, the printed lines should be clear and stable to produce a standard morphology (Le-Bail, Maniglia, and Le-Bail 2020). Lipton suggested that in extrusion printing, the material with shear thinning can better support itself after deposition. Formula $G^* = G' + iG''$ can express the influence of pressure on material flow performance, where G^* is the shear modulus indicating the shear thinning of the material, G' is the shear storage modulus, and G'' is the shear loss modulus. Under the action of pressure, the modulus parameter value will change, if the value of G'' is higher than G', the printed material can be extruded smoothly (Compton and Lewis 2014; Lipton 2017). When the pressure gradient acts along the length, the shear stress in the capillary also presents a radial change, as shown in Equation (1), where τ_r is the shear stress, ΔP is the pressure gradient, l is the length, r is



Figure 4. Using SHASAM technique to print a sugar-based toroidal coil sculpture, available at https://candyfab.org/.

the radial position within the nozzle. It can be seen that the shear stress on the central axis of the nozzle is zero, while the shear stress on the wall of the nozzle reaches its maximum value (Lewis 2002; Smay, Cesarano, and Lewis 2002).

$$\tau_{\rm r} = \frac{r\Delta P}{2l} \tag{1}$$

Godoi et al. suggested that an ideal extrusion printing material should also have an excellent performance of yield stress (YS), which includes the pressure required when flowing out of the static state and the pressure required to continue flowing in the motion state. This determines whether the printed material can pass through the nozzle, and whether it can withstand high shear rate and maintain minimum deformation (Godoi et al. 2019). Moreover, Godoi et al. also showed that moderate material viscosity is critical in the extrusion printing process of soft materials, which is too high to be extruded easily and too low to support the structure after deposition. If the viscosity of the material used for extrusion printing is not suitable, the rheological properties can be improved by adding additives or rheological modification (Godoi, Prakash, and Bhandari 2016). Wang et al. investigated the possibility of fish surimi gel as materials of 3D food printing. The results showed that surimi often had high viscosity and could not be extruded smoothly from the nozzle. By adding a certain amount of sodium chloride, the viscosity of surimi could be improved to make it extruded smoothly and maintain its shape after deposition (Wang et al. 2018).

Selective sintering printing and characteristics of food materials required

In selective sintering printing, the material powder is first laid into a powder bed, then the sintered source is used to heat the bed along a preset route, melting the powder particles and forming a solid layer. The sintered powder forms the product component, and the unsintered powder remains in place to support the formed structure. After a solid layer is completely formed, a new layer of powder is laid down for sintering. This process is repeated until the 3D food is printed layer by layer (Awad et al. 2020; Fina et al. 2017). According to the different sintering sources applied, selective sintering printing can be classified as selective laser sintering (SLS) and selective hot air sintering and melting (SHASAM) (Sun et al. 2015). The advantage of selective sintering is that the printing is more free and fast, and there is no need for subsequent curing. Moreover, during the printing process, each sintered powder layer can be altered to print out the

food with different ingredients in each layer, so that more complex food can be made. The disadvantage is that the printing process is complicated due to the many variables involved (Sun et al. 2015).

Although selective sintering printing has been successfully applied to sintered metals, ceramics, polymers and other materials (Barakh Ali et al. 2019; Ruggi et al. 2020), it has not been widely used in the food field due to the limitations of material properties. At present, the technique is mainly used to print sugar or lipid based food materials, which have a low melting point and can be easily sintered (Sun et al. 2015). Furthermore, the materials of selective sintering printing are limited to powder, and some food materials such as fresh food are not suitable for this printing technique, because their characteristics cannot meet the printing requirements (Mantihal, Kobun, and Lee 2020). Windell Oskay and Lenore Edman developed the CandyFab 4000 in the year 2007, which uses the SHASAM technique to print pure sugar powder and melt it with hot air in the process (CandyFab 2020). Figure 4 shows the printing process of a toroidal coil sculpture, one of the first objects printed by CandyFab 4000. The compressibility and particle density of powder materials are the characteristics that need to be considered, which will affect the fluidity of powder, thus affecting the formation of the target pattern by laser or hot air sintering (Berretta et al. 2013). Particle wettability is another important property of powder materials. It affects the amount of binder absorbed and the volume of binder dispersion in the powder substrate, which are vital to the physical properties of the printed product (Mantihal, Kobun, and Lee 2020). In addition, Diaz et al. suggested that materials suitable for SLS technique should not decompose under laser irradiation, especially at their typical heating temperatures of 140-180 °C. The powder material used should also be free flowing, meaning that the poured powder does not agglomerate and has no tendency to stick together or adhere to the contact surface. A numerical limit requires that the angle of repose be less than 30°. The angle of repose refers to the angle between the flat surface and the cone formed by the poured powder. It can be used to measure the flow characteristics of the powder (Diaz et al. 2016).

Binder jetting printing and characteristics of food materials required

Binder jetting printing is a technique for constructing 3D objects layer by layer in powder bed by spraying liquid binder. In the printing process, a counter-rotating roller is used to disperse each layer of powder material. When a

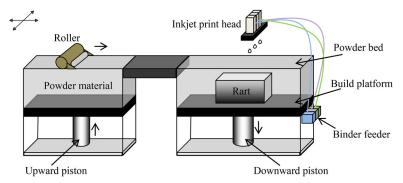


Figure 5. Schematic diagram of a Binder Jetting 3D printer.

layer of powder is ready, the inkjet print-head ejects the binder on a predetermined track to construct a 2D pattern on this layer of powder. The finished layer is then lowered along with the supporting platform to make room for the next layer of powder to disperse, and the printing process is repeated until the 3D target is formed (Holland et al. 2018; Ziaee and Crane 2019). Figure 5 shows a schematic of binder jetting equipment. Binder jetting printing can be used to quickly print products with inexpensive materials. However, the appearance of printed products is often not smooth enough. Moreover, the need for post-processing to solidify the product, and the high cost of required printing equipment, etc., are the factors limiting the use of this technique (Sun et al. 2015).

Materials for binder jetting printing are also limited to powders (Miyanaji et al. 2020). Mostafaei et al. suggested that powder characteristics have a significant impact on the printing process, and it is necessary to fully understand the characteristics, including the shape, appearance, size, distribution, fluidity, and accumulation density of powder particles. When the liquid binder drops into the powder bed, different powder morphology and particle size will affect the penetration rate of the binder. Typically, uneven powder distribution and excessive pore size increase the penetration time (Mostafaei et al. 2020). Powder fluidity is another key factor affecting the uniform distribution of powder on the powder bed. Powder materials with good fluidity can expand into a thin and uniform layer under the action of the counter-rotating roller. On the contrary, the powder material with poor fluidity is not easy to be spread evenly by the rotating roller (Holland, Foster, and Tuck 2019). Packing density is also an important characteristic of powder material, which is important for sintering and mechanical strength of products. High density of powder accumulation, internal void will be less, the sintered product porosity is lower, the final product mechanical strength is often higher. High packing density powders have fewer internal voids and the porosity of sintered products is lower, which makes the final printed products tend to have higher mechanical strength (Mostafaei et al. 2020).

Inkjet printing and characteristics of food materials required

Inkjet printing is a process in which droplets are produced under the guidance of a preset program and drop onto the

substrate to gradually form a 3D pattern (Kiefer and Breitkreutz 2020). According to the difference of droplet production process, the common inkjet printing technology mainly includes continuous inkjet printing (CIJ) and drop on demand printing (DoD) (Hoath 2016). In CIJ printing, the material is extruded through a nozzle in the form of a continuous flow of liquid, which are then dispersed into droplets under surface tension. There is usually a piezoelectric transducer in the nozzle device, and the liquid flow through the nozzle can be adjusted by changing the output frequency of the transducer, so as to enhance the dispersion degree of liquid materials. The dispersed droplets are generally induced to generate an electric charge. Under the action of an applied electric field, the charged droplets will move in a specific direction, so as to guide the extruded liquid material to drop in a set position and finally produce the desired print pattern (Daly et al. 2015). In DoD printing, the printing equipment often has multiple nozzles, and the printing material drops out from different nozzles as required, without the formation of liquid flow. The power source of droplet extrusion is generally generated in two ways: one is to install a piezoelectric ceramic element in the print head to generate power through its deformation (Huang et al. 2020), the other is to heat the liquid in the nozzle to expand it and squeeze it out of the nozzle through an electric heating element (Godoi et al. 2019).

The material characteristics required for inkjet printing are generally low viscosity so that they can be dispersed in droplet form after extrusion of the nozzle (Godoi, Prakash, and Bhandari 2016). This also prevents the technique from being used to print food with complex structures. Inkjet printing technique is now mainly applied to decorate and cover food substrates, as well as to fill material cavities. Butter, cream, chocolate, jam and other food materials are often used as food materials for deposition. FoodJet is a typical example of the commercial application of inkjet printing technique, with many accurate deposition products produced, as shown in Figure 6 (FoodJet 2020). Shastry et al. showed that the range of viscosity of liquids suitable for continuous inking is very narrow, with the average suitable inks having a viscosity of about 2.8-6 centipoise (cp). In the process of inkjet printing, the viscosity value above 10 cp is prone to cavitation, and below 2 cp is often unstable (Shastry et al. 2004). In addition to low viscosity, physical properties such as surface tension and inertia of liquid







Figure 6. Some deposit products produced by FootJet using ink-jet printing technique: (a) FoodJet marmalade depositor, (b) cavity filled biscuits, (c) cheese depositing on buns. The images available at https://www.foodjet.com/.

materials can also affect the printing effect. Daly et al. found that the Ohnesorge number (Oh) could be used to evaluate the performance of the droplets ejected from the nozzle, which is related to the density (ρ) , viscosity (η) , surface tension (γ) and size (d) of the liquid, as shown in formula 2:

$$Oh = \frac{\eta}{\sqrt{\gamma \rho d}} \tag{2}$$

If the Oh value is lower than 0.1, more satellite droplets will form in addition to the main droplets, and the liquid is too dispersed. Therefore, an appropriate combination of the properties of the liquid material is required to produce an ideal Oh value (Daly et al. 2015).

Novel evaluation technology of food material performance in 3D printing

Appropriate material characteristics are crucial for different 3D printing techniques. Therefore, it is necessary to evaluated the material characteristics prior printing and performance in 3D food printing. There are always some routine tests carried out to determine the properties of food material. For example, the rheological properties of the material will be determined by a rheometer, the texture properties will be measured by a texture analyzer, the powder properties will be analyzed by SEM and XRD, etc. In recent years, with the development of 3D printing technology, more and more engineering evaluation technologies suitable for 3D printing process have been introduced, which makes the selection and application of materials more advanced and easier. Table 3 shows the novel engineering evaluation technology for the characteristics of food materials in the 3D printing process.

Rapid evaluation of food material printing performance based on LF-NMR detection technology

Liu et al. introduced a method based on low field nuclear magnetic resonance (LF-NMR) parameters to quickly predict the 3D printing characteristics of mashed potatoes, which can evaluate the printing suitability of specific materials without carrying out tedious printing experiments (Liu, Zhang, and Ye 2020). The principle of LF-NMR technology is mainly based on the spin relaxation characteristics of hydrogen nucleus (oil/gas/water) in magnetic field to achieve rapid, accurate and nondestructive detection (Sun et al.

2019). It has been widely used in the field of food, such as monitoring the distribution and migration of water molecules in food drying (Li, Zhang, and Yang 2019; Sun, Zhang, and Yang 2019; Sun et al. 2021; Wang et al. 2021; Yang et al. 2020). At present, like rheological properties and texture analysis, LF-NMR is often used as a conventional analysis method in 3D food printing because the presence of water in a material is also an important property (Wang et al. 2018; Yang et al. 2018). Moreover, because of the close correlation between some physical properties of food materials and internal moisture, it is possible to indirectly reflect the physical properties of food materials by using LF-NMR to determine the distribution and migration of moisture in food materials, which is a higher level application of LF-NMR technology. This method has been successfully used to predict the dielectric properties of yam slices in drying (Li, Zhang, and Yang 2019), the quality of frying oil in ultrasonic assisted microwave vacuum frying (Sun, Zhang, and Fan 2019), and so on. Hence it is reasonable to consider the use of LF-NMR in the prediction of material properties in 3D food printing, since the properties of materials, especially the rheological properties which are crucial for printing effect, are closely related to moisture. In the study of Liu et al., a series of operations were carried out to achieve the purpose of using LF-NMR parameters to predict 3D printability of mashed potatoes, including (1) determination of rheological properties and printing properties of mashed potatoes with different formulations, (2) correlation analysis of rheological properties (G', G'', G^* , K, n) and printing performance by principal component analysis (PCA), (3) determination of the feasibility of using the rheological properties to evaluate the 3D printing performance by Fisher discriminant analysis, (4) establishment of correlation between LF-NMR parameters and rheological properties of mashed potatoes (Liu, Zhang, and Ye 2020). Sun et al. used LF-NMR technology to intelligently evaluate the effect of microwave pretreatment on the rheological property and 3D printing performance of the dough, considering that microwave treatment would further gelatinize the dough starch and change the water distribution. The Pearson correlation analysis showed that the rheological parameters and LF-NMR parameters of the dough were closely correlated. Then the prediction model based on LF-NMR and Partial least squares regression (PLSR) was established for the rapid detection of dough printing performance. The results revealed that the prediction of the model was stable and accurate, and the accuracy was over 90% (Sun, Zhang, and

Table 3 Novel engineering evaluation technology for the characteristics of food materials in the 3D printing process.

Method	Principle	Purposes	Application Target	Advantages
LF-NMR evaluation	Indirectly reflect the physical properties of food materials by using LF-NMR to determine the distribution and migration of moisture in food materials	Printing properties of food materials such as rheological properties	Printing materials	More convenient detection technology to predict food performance
Numerical simulation	Numerical simulation techniques such as computational fluid dynamics (CFD) are used to simulate the flow performance of materials	Prediction of food flow performance in extrusion printing process	Printing process	Accurate, simple and rapid evaluation method for extrusion-based 3D food printing materials
Simulated food system	Different types of water- based gels are used as simple reference models for foods to simplify evaluation	The printing performance of food can be evaluated by directly comparing with the characteristics of the reference material	Alternative reference materials	Small number of ingredients, mainly water and gels, it can be used to systematically study and understand the physical properties of internal structures that are not readily available in real food
Morphological identification	Capture views and record printout parameters as a basis for assessing material geometric properties and printing defects	Quantitative evaluation of geometry and defects of printed food by morphological identification	Printing process	The printing process can be directly recorded for evaluation, and the data obtained can also be used in numerical simulation

Chen 2020). Phuhongsung et al. also found the rheological properties of soybean protein isolate gel were strongly correlated with LF-NMR parameters, thus the LF-NMR spectroscopic characteristics could be used to predict the printing performance of soybean protein isolate gel (Phuhongsung, Zhang, and Devahastin 2020). Chen et al. compared the performance of LF-NMR and dielectric spectroscopy in predicting the rheological properties and 3D printability of surimi gels. The experimental results showed that the application of LF-NMR discriminant analysis display higher classification accuracy of printing performance, and the partial least squares regression (PLSR) model based on LF-NMR can demonstrate better prediction accuracy and stability (Chen, Zhang, and Yang 2021).

Rapid evaluation of food material printing performance based on computational simulation

Computational fluid dynamics (CFD), which utilizes computers to build mathematical models to simulate fluid flow, is widely used in many numerical simulation processes in the food industry, such as drying, refrigeration, sterilization, baking, cooking, mixing (Ghani et al. 1999; Norton and Sun 2006; Scott and Richardson 1997). In addition to predicting fluid flow behavior, CFD can also be applied to predict chemical reactions, mechanical motions, phase transitions, heat and mass transfer in food processing. The Navier's stokes transport partial differential equations is often used to describe the flow of fluid, taking into account the fluid mass, momentum and energy parameters, and adding other thermodynamic algebraic equations such as the density state equation and the constitutive equation to describe the rheological properties (Kuriakose and Anandharamakrishnan 2010). The application of CFD will help to solve the

equations, so that the complex physical mechanisms governing the thermal, physical and rheological properties of food materials can be understood more comprehensively and rapidly (Xia and Sun 2002). Numerous researches on CFD application focus on numerical simulation of food extrusion process to understand the fluid characteristics inside the extruder (Dhanasekharan and Kokini 2003; Emin and Schuchmann 2013; Sarghini, Romano, and Masi 2016; Singh and Muthukumarappan 2017). As material extrusion process also exists in 3D food printing, especially in extrusion printing technique, it is reasonable to believe that CFD can be applied in the prediction and evaluation of material characteristics in food 3D printing.

Guo et al. compared two different extrusion-based 3D food printing methods based on screw and syringe respectively in terms of fluid flow characteristics and printing profile through CFD and real printing experiment. The computational simulation model showed that the fluid characteristics of the material used in syringe-based printers during extrusion were simple, while that of the material used in screw-based printers during extrusion was relatively complex, with fluid backflow in the internal void. The CFD successfully evaluated the fluid characteristics of the two extrusion-based printing methods, which is consistent with the results of real printing experiment, indicating that CFD has the potential to predict the food material performance of 3D food printing (Guo, Zhang, and Bhandari 2019). By using computational fluid dynamics simulation technology, Guo et al. also developed an accurate, simple and rapid evaluation method for extrusion-based 3D food printing materials. First, five common grain gels were selected as test materials to determine their dynamic rheological properties and static rheological properties respectively. Since the grain gel tested was a shear diluted non-Newtonian fluid, Bird-Carreau model was introduced to describe the dynamic

rheological properties of the system, so as to obtain more realistic simulation results. These dynamic parameters were then applied to CFD simulation. Finally, the simulated pressure distribution value can be used to evaluate the extrudability of the material, because it determines whether the material in the syringe is subjected to appropriate pressure. Only under appropriate level of pressure can the material in the tube be successfully extruded. The higher the simulated pressure value is, the more difficult it is to extrude the material, which has been proved by experimental results (Guo, Zhang, and Devahastin 2020).

There are some other computational simulation methods have been used to evaluate the performance of 3D food printing. Yang et al. studied the effect of the addition of different starch on the printing performance of lemon juice gel. In addition to the influence of different starch components, the rheological properties of lemon juice varied before and after gelation and under different printing parameters, but it is difficult to detect the dynamic rheological properties of the material due to the sealing property of the printer. Therefore, as a powerful simulation tool, POLYFLOW software based on finite element method was used to simulate the fluid characteristics of lemon juice in the flow channel of 3D printer under different process parameters (Yang et al. 2019). Liu et al. used numerical simulation to evaluate the dynamic extrusion 3D printing process for mixed fluids. POLYFLOW software was applied to calculate the viscosity and pressure distribution of the fluid during extrusion and describe the relationship between the rheological properties, flow field distribution and printing characteristics. Finally, the prediction and evaluation of the 3D printing performance of mixed fluid materials can be achieved through the established relationship (Liu et al. 2020). Jonkers et al. established a constitutive model applied in finite element to predict the mechanical properties of 3D printed food. The influence of geometric effect was illustrated by the simulation, and the mechanical properties of printing materials can be evaluated by identifying model parameters (Jonkers, van Dommelen, and Geers 2020). Duty et al. developed a material evaluation model based on fluid change and thermophysical properties for various extrusion printing methods. The model applied equation parameters to define the material characteristics that meet the printing requirements. It is found by sample test that the model evaluation results are consistent with the experimental results, which indicates that the model can successfully evaluate the material characteristics for 3D food printing (Duty et al. 2018).

The quality of the final printed 3D food can be monitored by shape fidelity, which refers to the degree to which the printed and maintained shape is close to the preset 3D model. Nijdam et al suggested that there are three factors contributed for shape fidelity, including printer capability, filament quality, and dimensional stability (Nijdam et al. 2021). They further proposed a method to screen food printing materials with sufficient rigid support for threedimensional structures. The storage modulus and damping factor extracted from the food material amplitude sweep were used to form the axis of a graph, and a dimensional

stability window is defined on the graph by setting the limit change parameters. This window allows the stability of 3Dprinted food to be predicted by simply testing the rheological properties of the food material, thus avoiding the need for arduous printing tests to determine the suitability of the material (Nijdam et al. 2021).

Evaluation of food material printing performance based on alternative reference material

It is challenging to evaluate the printing characteristics of food materials under different printing parameters due to their complex composition and changeable material properties. Different types of water-based gels can be considered as simple reference models for food. Due to its small number of ingredients, mainly water and gels, it can be used to systematically study and understand the physical properties of internal structures that are not readily available in real food. Many hydrocolloids with different molecular weight and structure exist in natural plant cells, which provides the possibility to simulate the real food fluid characteristics (Gholamipour-Shirazi, Norton, and Mills 2019; Pegg 2012; Vilgis 2015). Kim et al. established a method to systematically evaluate the printing performance of selected foods using hydrocolloids as a reference material, and classified selected foods according to the evaluation results as a standard, which can be used as a basis for selecting raw materials for 3D food printing. Methylcellulose was used as the reference material and 12 kinds of food with different rheological properties were selected to verify the simulation results. The experimental results showed that the selected reference materials can completely simulate the deformation behavior and processing characteristics of real food, and the printing performance of food can be evaluated by directly comparing with the characteristics of the reference material (Kim, Bae, and Park 2018). A similar approach has also been found in a paper published Xu et al., in which a small molecule drug indomethacin was used as a reference model compounds. The drugs mixed with polymers with different solubility were used as extrusion based 3D printing materials. By analyzing the relationship between mechanical properties and printing properties of extruded filaments and identifying the parameters of "toughness," etc., a method for quantitative evaluation and prediction of printing properties of filaments for fused deposition modeling 3D printing was developed (Xu et al. 2020).

Evaluation of food material printing performance based on morphological identification

Literature indicates that in addition to numerous studies focusing on the evaluation of printing performance of materials through their rheological properties, there are also some studies involving quantitative evaluation of geometry and defects of printed product by morphological identification. The product geometry and defects can directly reflect the printing performance of food materials. Fahmy et al. developed a camera-based morphological identification

method to evaluate the printing quality of starch-based materials. The raw material was a mixture consisting of wheat flour with different water content and wheat starch/ egg white protein, which were used to simulate a series of properties of printing materials and their interactions with the printing process. Two cameras were used to capture views from different angles and record printout parameters as a basis for assessing material geometric properties and printing defects. This is the first time to use an on-board camera to quantitatively study the dimensional attributes of printing product and the performance of food materials in 3D printing. The research results can also be used to construct the prediction model of the printability of viscoelastic food materials for 3D printing (Fahmy, Becker, and Jekle 2020). It is worth noting that when Liu et al. studied the correlation between rheological properties and printability of the carrageene-xanthan-starch gel system in extrusion 3D printing, they adopted a non-automatic geometric shape assessment method based on camera system, but did not consider the overall geometric shape stability and printing defects (Liu et al. 2019).

In the Inkjet printing technique, the droplet injection behavior dominated by the fluid flow pattern is critical to deposition quality of final product, so the evaluation of the fluid material flow pattern is important to improve the Inkjet printing effect. However, the current static image evaluation method has obvious drawbacks, which cannot record the evolution process of droplet in time. To solve this problem, Huang et al. used a camera in Inkjet printing technique to collect dynamic video data to evaluate the evolution of droplets. The video capture device consists of a sensor camera equipped with magnification lens and a stroboscopic LED for illumination, which collects droplets' dynamic date by adjusting the delay time between the jetting signal and the LED lighting signal, as shown in Figure 7. In addition, they also proposed an unsupervised learning method based on deep recurrent neural network (DRNN) technology to study droplet flow patterns in video. Through simulation and experimental data verification, this method can accurately express dynamic video data, which is of great value for the evaluation and prediction of droplet behavior (Huang et al. 2020).

Some novel instrumental analysis techniques for evaluating food material printing performance

In addition to the traditional 3D printing product characterization technology, some instrumental analysis technologies that are not usually used in 3D printing have been introduced recently. When Liu et al. studied the structure and rheological properties of potato starch during hot extrusion 3D printing, Attenuated total reflectance-fourier transform infrared spectroscopy (ATR-FTIR) was used to analyze the short-range order of the potato starch. The results showed that hot extrusion can damage the short-range ordered structure, and the destruction of short-range order is mainly affected by the printing temperature, not by the starch concentration. In order to evaluate the submicroscopic structure

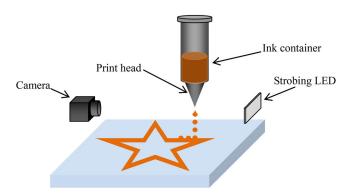


Figure 7. Schematic diagram of Inkjet printing with data collection devices.

of printed products, Small-angle X-ray scattering (SAXS) was used to analyze the changes of crystal structure inside potato starch. The results confirmed that hot extrusion printing destroys the original starch crystal form and promotes the formation of new gels with ordered network structure (Liu et al. 2020).

In the printing of polymer materials, because the 3D target is produced layer by layer, it is necessary to keep the molten state when the new layer is deposited, so as to effectively bond with the bottom layer. Moreover, the temperature should not be too high, otherwise the material viscosity is too low, and the printed parts will be deformed or even collapse under their own weight. Therefore, the best condition for printing is to keep the temperature slightly above the glass transition temperature of the material throughout the process, rather than just at the moment the material is extruded. In order to achieve this goal, Dinwiddie et al. used medium wave and long wave infrared cameras to measure the temperature distribution in the extrusion printing process of thermoplastic materials, and further made a quantitative analysis of the relationship between temperature and zstrength (force perpendicular to the layer plane) in the deposition process (Dinwiddie et al. 2014).

Future scope

Diversified design, personalized customization, complex structure printing and material saving are the advantages of 3D food printing. With the increase of population in the future, food crisis will be prominent. The promotion and application of 3D printing can reduce food waste and contribute to global food security. It is believed that 3D food printing technology will be more and more widely applied. However, few of the 3D printing products that have been industrially produced today involve people's nutritional staples such as rice and vegetables, because these raw materials are not suitable for direct printing. Therefore, it is necessary to develop better evaluation technology for 3D food printing, and the evaluation results can be used as the basis for selecting materials or adjusting the material formula. In fact, there are many kinds of food raw materials, and the complex and changeable composition of the materials brings challenges to the evaluation work. Although there are some traditional evaluation methods, they are not systematic and the results often cannot fully reflect the characteristics of



food materials. The evaluation methods of 3D printing materials should be standardized to meet the detection requirements of different materials.

In addition, in the process of 3D printing, there are many factors affecting the material performance, such as temperature, speed, shear force, etc., and the material printing characteristics are also in the process of dynamic change. Some studies have found that the rheological properties of materials will change with time (Sun et al. 2018). Therefore, the performance of food materials in printing as a constant value to test is often not appropriate. Using mathematical model simulation to obtain dynamic values is a good solution, which is also another development direction of 3D printing evaluation.

On the other hand, due to the edible characteristics of food products, people should pay more attention to the microbial safety and food safety in the whole printing process, and the evaluation content should also include the detection of microorganisms and the evaluation of food safety.

Conclusion

It is found that although there are some native printable foods, the raw materials that can be used for 3D printing are limited due to unsuitable material properties. For nonnative printable food, its printing characteristics can be improved by adding additives, but the premise is to understand the printing technology's demand for material characteristics, so as to adjust the food formula purposefully. It can be concluded that the characteristic of the material is the key factor determining the success of printing. At present, there are mainly four techniques used for 3D food printing, each of which has different requirements for material properties, but there are also some commonalities. Generally, food materials are required to have good rheological properties for easy extrusion and suitable mechanical properties to maintain shape after deposition. This requires the corresponding detection technology to evaluate the material characteristics and food printing performance. On the basis of some existing traditional evaluation techniques, more and more new evaluation techniques have been introduced, including computer dynamic simulation, low field nuclear magnetic resonance indirect evaluation, product morphological identification, etc., making the evaluation techniques for 3D food printing more and more accurate and comprehensive.

Disclosure statement

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