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REVIEW



3D printing of food: pretreatment and post-treatment of materials

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ABSTRACT

Background: Food 3D printing is an emerging food processing technology. Due to the advantages of functionalization, customization, personalized nutrition design, simplified supply chain and broadening existing food materials, 3D printing has been extensively studied in the food sector in the past decade. Many factors influence the accuracy and quality of food 3D printing, which are also the challenges to researchers. Currently, most of the research focuses on the development of printable materials and control of printing parameters to improve the printing accuracy and product quality. However, the influence of material pretreatment methods and post-processing techniques on food 3D printing have received less attention.

Main content: By collecting the available data and research, this paper analyzes the effect of pretreatment technologies (crushing, gelation, etc.) and post-treatment technologies (cooking, drying, fast cooling technology, 4D printing, etc.) on the accuracy and shape fidelity of 3D printed food products. It also summarizes the current challenges of food 3D printing and proposes some thoughts on the future development of this technology.

KEYWORDS

3D printing; Foods; Pretreatment; Post-treatment; Printing precision; Shape fidelity

Introduction

3D printing, also known as additive manufacturing technology (AM), rapid prototyping technology (RP) or solid free-form fabrication (SFF), is a technology based on digital models to produce pre-designed 3D objects by layering adhesive materials (Dankar et al. 2018; Godoi, Prakash, and Bhandari 2016; Liu et al. 2017). Different from traditional manufacturing methods, 3D printing technology has been evolved tremendously in recent years due to its potential advantages, such as customized geometry, reduced production cost, shortened manufacturing cycle and almost unconstrained complexity of appearance, which have been regarded to potentially promote the third industrial revolution (Liu et al. 2017; Sun et al. 2018; Wang et al. 2017). Currently, 3D printing technology has been applied in medical, industrial manufacturing, biological tissue, architecture, aerospace, education, clothing, and packaging sectors (Abouhashem et al. 2015; Murphy and Atala 2014; Vanderploeg, Lee, and Mamp 2017; Yang, Zhang, and Bhandari 2017), but the application in food industry is still in the infant stage. Yang et al. (2001) has been reported to be the first time using the extrusion based technology to produce a complex 3D cake. Since then, 3D printing technology has gradually developed in food sector.

With the increased living standards, people have higher demand on healthy functional food and even personalized food (Sun et al. 2018). According to a 2015 survey, most consumers considered themselves as “health-conscious” or

“ingredients sensitive” (Deloitte 2015). Food 3D printing can be blended with various raw materials according to individual's physical and nutritional status, so that the functional factors such as protein, fat, dietary fiber, vitamins, and minerals, are balanced according to the demand (Dankar et al. 2018; Feng, Zhang, and Bhandari 2018; Liu, Zhang, and Bhandari 2018; Sun et al. 2018). In addition, it can also broaden the source of food materials such as algae (An et al. 2019) and edible insects (Severini, Azzollini et al. 2018) in food processing, and realize digital nutrition and complex food design (Liu, Bhandari, Prakash, and Zhang 2018), which cannot be achieved by traditional food processing methods. For example, the 3D printed foods were much tender and had personalized nutritional compounds which were specifically produced for the elderly and patients who have difficulty in swallowing (Kira 2015; Kouzani 2017; Lunardo 2016). Derossi et al. (2018) also developed a fruit-based children snack using 3D printing technology, which provided 5-10% of the daily recommended intake of energy, vitamin D, iron, calcium and other nutrients for children aged 3-10 years. The success of these examples make it possible to customize meals for special group of people such as army, athletes, astronauts, and pregnant women.

Currently, there are four types of 3D printing technologies for food processing: extrusion-based printing, selective laser sintering, binder jetting and inkjet printing (An et al. 2019; Izdebska and Zolek-Tryznowska 2016; Liu et al. 2017). Among them, extrusion-based 3D printing is the most

widely used, which can be further classified into room temperature extrusion, fused deposition manufacturing and gel-forming extrusion, according to different printing material states (Godoi, Prakash, and Bhandari 2016; Liu, Bhandari, Prakash, Mantihal, et al. 2019; Sun et al. 2018). Room temperature extrusion is carried out without temperature control and phase transition, and the extrusion of dough falls into this category (Yang, Zhang, Prakash, et al. 2018). To achieve successful printing, appropriate viscoelasticity of the material is the key property, which allows the material to be extruded through a fine nozzle and support the product structure after deposition. For melt extrusion molding, the molten paste is extruded from a nozzle and almost immediately solidifies and is welded to the previous layer, and this technique has been widely used to create customized 3D chocolate products (Liu et al. 2017; Mantihal et al. 2017). Different from the former two types of extrusion molding, the application of gel extrusion molding is much broader, such as the printing of hydrogel (Cohen et al. 2009), meat (Wang et al. 2017), mashed potatoes (Liu, Zhang, and Bhandari 2018; Liu, Zhang, et al. 2018), cheese (Kern, Weiss, and Hinrichs 2018), and fruits and vegetables (Severini, Derossi et al. 2018). In contrast, selective laser sintering (SLS) is a technique in which power lasers are used to selectively fuse powder particles into a three-dimensional structure layer by layer. The laser scans the cross section of each layer surface to selectively fuse the powder. After scanning each section, the powder bed is lowered and covered with a new layer of powder, and then repeat this process until the desired structure is completed. Finally, the untreated powder is removed and recycled for the next operation (Liu et al. 2017; Noort et al. 2017). The SLS is widely used in the manufacture of cermet industry, but there are certain restrictions in the application of food because it is only used for the printing of powdery ingredients with low melting point (chocolate powder, sugar-based materials, fat-based materials, etc.) (Diaz et al. 2016; Godoi, Prakash, and Bhandari 2016; Liu et al. 2017; Sun et al. 2018). However, it is interesting to note that the method is completed by layer-by-layer laying of powder, so the powder of each layer can be different, which is beneficial to the balanced processing of foods (Liu et al. 2017). Binder jetting is similar to SLS in principle. In the printing process, layers of powdery material are deposited, and the binder is selectively sprayed onto each layer of material in a specific area. The binder melts the current section onto the sections before and after the weld. In the manufacturing process, the unmelted powder always supports the molten parts, resulting in complex structures. Finally, the unbonded powder is removed and recycled (Sun, Zhou, Huang et al. 2015). Binder jetting has the advantages of fast processing speed and low cost, but the smoothness is not enough, which requires post-treatment operations such as high-temperature curing (Sun, Peng, Yan, et al. 2015; Sun, Peng, Zhou, et al. 2015). This technique usually fabricates food using sugar and starch powders. Inkjet printing uses a drop-to-drop method to print food. Compared with the previous methods, it is commonly used for surfaces filling or image decoration

on food surfaces (Liu et al. 2017; Pallottino et al. 2016), suitable for printing of low-viscosity materials such as pulp or paste (Liu et al. 2017; Sun, Peng, Yan, et al. 2015).

The recent research in food 3D printing technology is mainly focusing on exploitation of 3D printing materials and optimization of instrument processing parameters, which are important to the success of food 3D printing. However, the research on pre-/post-processing technology of 3D printed materials have received less attentions, which are also critical for the successful printing. Therefore, the objective of this paper is to analyze the effects of pretreatment and post-treatment techniques on the printing accuracy and shape fidelity of 3D printed food products. The challenges and future development of food 3D printing are also discussed.




Pretreatment of food 3D printing materials

For 3D printing, it is important to select materials with appropriate physical and chemical properties, such as particles size, fluidity, rheology, and mechanical properties. Initially, the materials commonly used for 3D printing are metals, ceramics, cells, tissues and synthetic polymers, which are carried out in organic solvents, crosslinking agents and extreme conditions, and do not meet food safety standards. Therefore, the selection of food-grade materials has become one of the big challenges in 3D printing of food products. The food materials should have appropriate fluidity, viscosity, rapid recovery performance and proper mechanical strength, so that they can easily flow out from the nozzle tip and are capable of self-supporting and maintaining the shape after printing. Therefore, pretreatment of food ingredients to meet these properties, like proper fluidity, rapid recovery behavior and appropriate mechanical strength, are very critical to achieve successful printing. At present, food 3D printing materials can be divided into three categories: powder, gel system and “dough” (Table 1).

Preparation of powder materials

Powdered materials are widely used in the above four types of 3D printing, but each technique has some specific requirements on the powder properties (Holland et al. 2018; Holland, Tuck, and Foster 2018; Liu et al. 2017). For example, fused deposition manufacturing requires the powder with a lower melting point, a reasonable cooling rate, appropriate particle size, viscosity and fluidity to facilitate the extrusion molding of the nozzle and maintain the stability of the shape after deposition (Godoi, Prakash, and Bhandari 2016), such as in printing chocolate (Hao et al. 2010; Mantihal et al. 2017). However, in addition to the proper particle size and flowability, SLS and binder jetting printing require the powder with certain level of bulk density and wettability (Godoi, Prakash, and Bhandari 2016; Liu et al. 2017; Shirazi et al. 2015). To meet the material requirements of various printing technologies, it is often necessary to pretreat the materials before printing, such as comminution and microencapsulation.

Table 1. Comparison of different material states during 3 D printing.

	Powder	Gel system	Dough
Printing technology	Fused deposition manufacturing; Selective laser sintering; Binder jetting; Inkjet printing	Extrusion based printing; Binder jetting; Inkjet printing	Extrusion based printing
Material properties	Particle size, Viscosity, Melting point, Wettability, Flowability et al.	Rheological properties, Gel properties, Thermodynamic properties	Viscosity, Mechanical strength, Flowability
Pretreatment technology	Comminution, Microencapsulation	ionotropic crosslinking, heat treatment gelation, enzymatic crosslinking, other methods	Formula optimization, Alternative ingredient, Gelatinization
Products	a. 	b. 	c. 

*The products images are obtained from the following websites: (a) Selective laser sintering of sucrose (Hong 2014); (b) Extrusion-based 3 D printed product of lemon juice gel (Yang, Zhang, Bhandari, et al. 2018); (c) 3 D printing of baked dough (Yang, Zhang, Prakash, et al. 2018).

Comminution

One of the most important purposes of comminution is to obtain the powder with appropriate particle size and flowability. This is the basic requirement for the material to flow out from the nozzle. Too large or small particles have a negative impact on 3 D printing. There are many methods for powder grinding, among which ball mill treatment is very common in food 3 D printing. It uses impacts of falling abrasive bodies (such as steel balls, goose hatching stones, etc.), the grinding action of the grinding body and the inner wall of the ball mill to crush and mix materials. Studies have shown that ball milling can transform crystalline cellulose into an amorphous state and recrystallization under suitable conditions to form a rigid structure (Abbaszadeh, Macnaughtan, and Foster 2014; Avolio et al. 2012). Holland et al. (2018) used ball-mill treatment on cellulose powder and found that a small amount of xanthan gum mixed with cellulose can synergistically enhance bonding properties and improve the inkjet printing performance. After the cellulose was mixed with locust bean gum or konjac in a ratio of 9:1 and then ball milled, squares of 10 mm × 10 mm shape were successfully printed (Holland, Tuck, and Foster 2018). In addition, other researchers showed that the neutral protease could be used to remove the connective tissue and fat of fresh meat. After freeze drying, coarse pulverization and ultrafine pulverization using a planetary ball mill, nano-meat powders were obtained which were suitable for food 3 D printing (Liu et al. 2017; Sun, Peng, Zhou, et al. 2015).

Microencapsulation

Microencapsulation technology is a technology that can form solid particles by embedding gases, solids and liquids in a microcapsule (Fang and Bhandari 2010). In food sector, it is often used to embed functional components such as polyphenols, enzymes, probiotics, functional oils, vitamins, minerals, etc. in capsules to protect against the impact of the

surrounding environment (Borgogna et al. 2010; Nazzaro et al. 2012). Studies have shown that the particle size of microcapsule powder could exhibit a bimodal distribution by controlling the microencapsulation conditions (Carneiro et al. 2013), which is ideal for binder jetting printing, where smaller particles can be filled between larger particle gaps to reduce porosity and improve mechanical strength. Therefore, microencapsulation could be a very promising pretreatment method for preparation of 3 D printing materials.

Only a few powdered materials such as powdered sugar, chocolate powder and some starches can be printed by fused deposition manufacturing, inkjet printing, selective laser sintering, and binder jetting. However, most of the powders are not directly used for food 3 D printing. They need to be mixed with suitable solutions to form soft materials such as gel, mud, and dough, before the 3 D printing operation.

Preparation of gel

Gels include dry gels, hydrogels, aerogels, and organo-gels depending on the dispersion medium (Gulrez, Al-Assaf, and Phillips 2011). In food sector, many high molecular polymers such as polysaccharides, peptides, and proteins hydrophilic polymers, so water is often used as dispersion medium to form hydrogels ('gel'). In recent years, extrusion-based printing of hydrogels is one of the main research topics in food 3 D printing. The printability of the gels is depended on the rheological (viscoelastic) and gelation mechanism of the polymer solution (Godoi, Prakash, and Bhandari 2016). The cross-linking methods commonly used to form the gels in food 3 D printing can be roughly classified into ionotropic crosslinking, heat treatment gelation, enzymatic crosslinking, and other methods.

Ionotropic crosslinking

Ionotropic crosslinking is a physical crosslinking method (Gulrez, Al-Assaf, and Phillips 2011; Pellá et al. 2018),

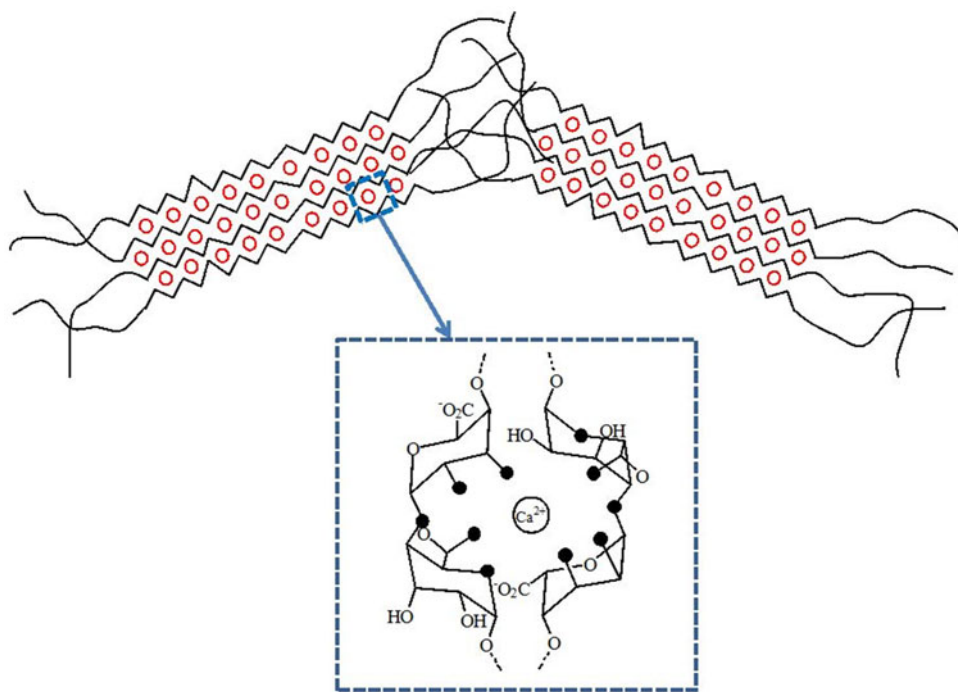


Figure 1. Structure of gel formed by alginate and calcium ions (Gulrez, Al-Assaf, and Phillips 2011).

forming a network structure of ionic cross-linking due to polyelectrolytes cross-linking with counter-ions. The most typical example is the preparation of sodium alginate hydrogel (Liu, Liu, et al. 2018). Sodium alginate is a natural anionic polymer composed of β -D-mannuronic acid (M) and α -L-guluronic acid (G) (Giuseppe et al. 2018), and the divalent cations can chelate with a plurality of oxygen atoms in the polymer to form hydrogel with a three-dimensional network structure (Fig. 1). Since divalent calcium ion (Ca^{2+}) is one of the most essential elements in the human body, it is currently the most commonly used cross-linking ion (He et al. 2016). It was reported that the calcium alginate gel microspheres formed by the cross-linking of alginate and divalent calcium ion (Ca^{2+}) were negatively charged, which could be deposited according to the established route under the driving of the electric field to complete 3D printing (Chen et al. 2018; He et al. 2016). Researchers mixed sodium alginate solution with mouse fibroblasts, and then rapidly gelatinized with 100 mM calcium chloride solution (Shi, Laude, and Yeong 2017). By changing the concentration of sodium alginate solution, 3D structures with different mechanical properties could be printed, and the cell activity were maintained (Shi, Laude, and Yeong 2017). Although this example is the application of calcium alginate gel in 3D printing of mouse cells, it provides an idea for printing functional foods such like probiotic foods.

Another gel formation material is pectin which is a heterogeneous polysaccharide polymer consisting of rhamnogalacturonan and homogalacturonic acid (Vancauwenberghe et al. 2017). The galacturonic acid can be methylated and classified as low methoxyl pectin (LM) and high methoxyl pectin (HM) according to the degree of methylation. Their gelation mechanisms are also different. HM is generally dissolved in low pH and high sugar solutions by hydrophobic

interactions and hydrogen bonding interaction to form gel, but LM forms gels by ion cross-linking between free carboxyl groups and Ca^{2+} (Fraeye et al. 2010), which is similar to that of alginate forming gel with Ca^{2+} . Vancauwenberghe et al. (2017) prepared a series of pectin-based gels by changing the LM, CaCl_2 , bovine serum albumin and sirup concentrations, and the result showed that for food gels without sirup addition, the gel with R value ($R = [\text{Ca}^{2+}]^2 / [\text{COO}^-]$) between 0.2 and 0.5 had a proper fluidity and stability for 3D printing; When $R > 0.5$, the printing performance was good, but the mechanical properties of the printed products were poor; In most cases, the addition of sugar increased the viscosity of the gel and had a positive effect on printing performance and molding quality. This research confirmed that pectin-based food inks can be used to print candy and other foods with variable microstructures and textures. However, since the gels after extrusion need to be incubated in CaCl_2 solution for some time to complete the gelation, 3D printing using dual nozzles could save the processing time. In a dual nozzle 3D printer, the inner nozzle is filled with low methoxy pectin solution, and the outer nozzle is filled with CaCl_2 cross-linking solution, which forms stable gel structures simultaneously during the 3D printing process. The product formed by this method does not require post-treatment, and the structure and shape of the printed food can be precisely controlled (Vancauwenberghe et al. 2018).

Heat treatment gelation

Heat treatment gelation is also widely used in food 3D printing. Different from ion crosslinking, heat treatment gelation mainly relies on the material sensitivity to temperature to form a gel. Many natural polymers or modified polymer

gel systems have certain heat sensitivity such as gelatin, starch, agar, carrageenan and cellulose derivatives (Vancauwenberghe et al. 2018). These thermo-reversible gels behave similarly. At higher temperatures they have a conformation of coils, but as the temperature decreases, they begin to form helical structures and aggregates, forming a gel. Many researchers have used this mechanism to print protein, potato starch, fruits and vegetables, and egg white protein. At present, the most widely used heat treatment method is water bath heating. For example, Liu, Meng et al. (2019) mixed the egg white protein solution with a mixture of gelatin, corn starch and sucrose at 55 °C for 10 min to prepare a 5% printing complex mixture system and stored at 40 °C for 20 min before printing. The results indicated that adding egg white protein can significantly improve the hardness and elasticity of the gel, increase printing precision and shape stability, suggesting the egg white protein multi-material system is a promising 3D printing material. Lille et al. (2018) mixed 30% rye bran, 35% oat protein concentrate or 45% broad bean protein concentrate, heated in a water bath, and then cooled to 4 °C to form a gel for 3D printing, which resulted in good printability with high product stability after printing. A paste-like substance by mixing whey protein isolate and milk protein concentrate in a ratio of 2:5 and adding a certain amount of glycerin and xanthan gum can also be successfully 3D printed (Liu, Liu, et al. 2018). In another study, κ -carrageenan, xanthan gum and potato starch were dissolved in water at a certain ratio and heated in a water bath at 90 °C for 30 min. After homogenization, the gel was cooled to 4 °C to form a gel for 3D printing (Liu, Bhandari, Prakash, Mantihal et al. 2019). The results showed that the addition of starch and xanthan gum to the carrageenan-based ink can improve the gelation property, reduce gelation time, and improve print performance and shape fidelity (Liu, Bhandari, Prakash, Mantihal et al. 2019). Kim et al. (2018) added different levels (10% and 30% w/w) of vegetable powder (broccoli, spinach, carrot) to xanthan gum solution and examined their printing properties (Fig. 2). The results showed that xanthan gum with its high hydration ability can inhibit the expansion of the vegetable powder particles, so that the rheological value of the gel system before and after the powder addition was minimally changed, and the difference of printability between different vegetable powders were reduced. Therefore, the gel system with different vegetable powders was smoothly extruded and had high printing precision and good stability. Azam, Min, et al. (2018) added 15 g of wheat starch and 1 g of different hydrophilic colloid mixture to 100 g of orange concentrate (OC), then the mixture was steam cooked for 20 min. After cooking, when the temperature of the mixture dropped to 40 °C, 1 ml of vitamin D was added and stirred for 5 minutes, and a gel enriched with vitamin D was formed for 3D printing. The results showed that carrageenan could bind to the double helix structure of amylose/amylose and improve gel printing performance and mechanical strength. Similarly, the authors also found that the gel formed by 20% wheat starch and orange concentrate had very good rheological and mechanical properties, and high

printing accuracy (Azam, Min, et al. 2018; Azam, Zhang, et al. 2018). Other researchers mixed concentrated strawberry juice (Liu, Zhang, and Yang 2018), lemon juice (Yang, Zhang, Bhandari, et al. 2018) with starch to form a gel for 3D printing. These studies demonstrated the feasibility of 3D printing of fruit concentrates and juices.

Compared with the traditional water bath heating, microwave heating has the advantages of fast heating, high energy efficiency, easy control and good sanitary conditions (Fu et al. 2012). Some researchers have used microwave heating to induce the formation of a surimi gel system and compared with the traditional water bath heating induced gel system. When preparing a surimi gel using conventional water bath heating, the surimi was heated in a water bath at 40 °C for 30 min and then heated in a water bath at 85 °C for 30 min. However, microwave heat treatment was only required for 60 s at 15 W/g power, and the surimi gel system has better mechanical properties and functional properties (Fu et al. 2012). Similarly, Ji et al. (2017) compared the effects of microwave heating and water bath heating on the polysaccharide protein gel system and found that compared with water bath heating, microwave heating could significantly reduce the heat treatment time, and the microwave heated gel has a denser network structure and a more uniform distribution of the polysaccharide network structure in the gels. This means that the microwave heating could be a more efficient method to induce gel in 3D printing.

Enzymatic crosslinking

Transglutaminase (TG) is an extracellular enzyme with a wide range of sources, low substrate specificity and low environmental sensitivity (such as heat and pH). It is a commonly used enzyme that induces the formation of protein gels. Transglutaminase can change the structure of protein, catalyzing the acyl-transfer reaction between the γ -carboxamide group of the glutamine residues and the ϵ -amino group of lysines in proteins, resulting in intermolecular or intramolecular crosslinking [ϵ -(γ -glutamyl)-lysine cross-linking], thereby promoting the formation of the protein gel (Dickinson and Yamamoto 1996; Wang et al. 2017). At the final process of a transglutaminase involved gel formation, the cross-linking reaction can be terminated by changing the temperature to inactivate the enzyme, which is simple and easy to control. Lipton et al. (2010) achieved 3D printing of meat by adding TG to minced turkey and scallop meat slurry, and the printed product could maintain a stable structure after cooking. Schutyser et al. (2018) prepared transglutaminase crosslinked sodium caseinate to investigate the printability. It was found that the gelation temperature of sodium caseinate was increased after the transglutaminase treatment (Fig. 3), which is a good property because even low concentration of sodium caseinate 25% (w/w) in this system was printable. Irvine et al. (2015) also established a method for cross-linking TG and gelatin to 3D printing of cells, which might be used to development of functional foods containing bioactive cells.

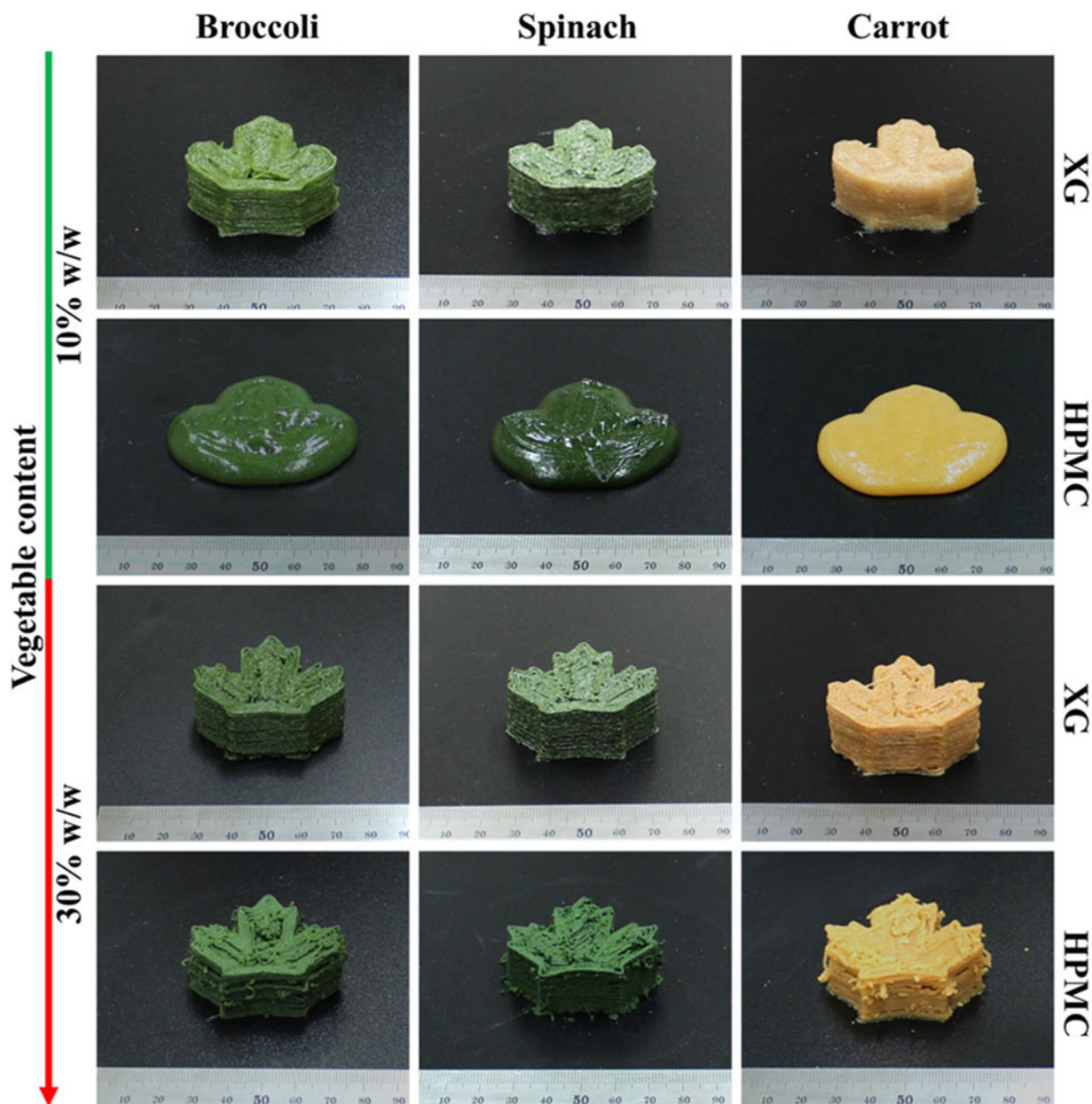


Figure 2. Image of XG and HPMC products containing 10% and 30% vegetable powder (Kim et al. 2018). (HPMC: hydroxypropyl methyl cellulose, XG: xanthan gum).

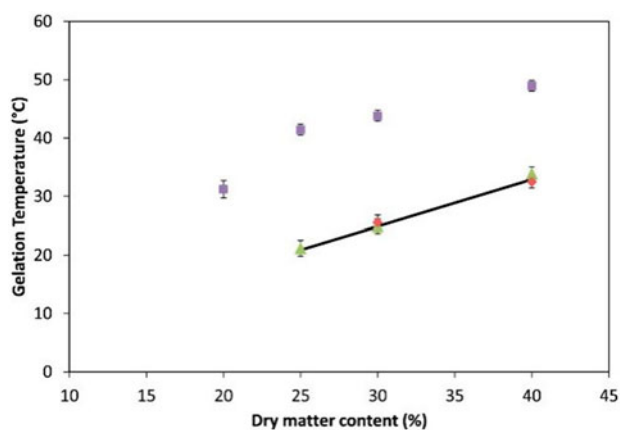


Figure 3. Changes in gelation temperature of different formulations gel (Schutyser et al. 2018). "Red diamonds" indicate the gel temperature at which no additives are added; "green triangles" indicate the gel temperature at which the additive is added; "blue squares" indicate the gel temperature at which the sodium caseinate is crosslinked with the transglutaminase.

Other methods

In addition to the above methods, the 3D printing gel system can also be formed by acid induction, complex coacervate formation and double crosslinking gelation.

There are two ways in which acid-induced gel formation can occur (Wang et al. 2017): (i) addition of an acid or acidulant in a protein solution and (ii) dialysis method, which achieves a slow pH reduction by dialysis in an acidic solution. Gluconolactone is a weak acid that is slow to acidify but easy to control in food systems (Totosa et al. 2002). When dissolved in water, it releases gluconic acid and slowly break down into hydrogen ions, which reduces the acidity of the protein solution and thus changes the microenvironment. At the same time, it weakens the electrostatic repulsion between protein molecules, increases their interactions, leads to protein aggregation and precipitation, thus forms a gel system. This acid induced gel system is

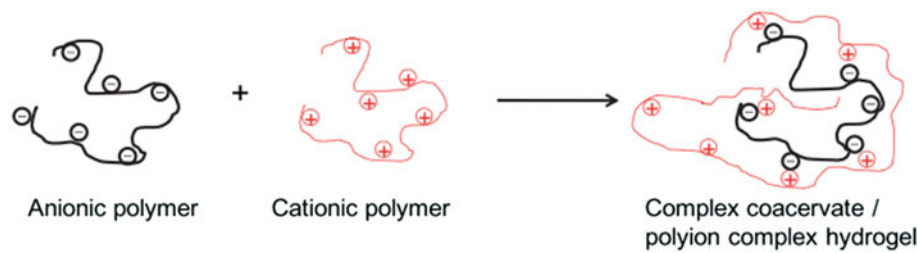


Figure 4. Complex coacervation between polyanions and polycations (Gulrez, Al-Assaf, and Phillips 2011).

suitable for extrusion-based 3D printing (Feng, Zhang, and Bhandari 2018; Wang et al. 2018). For example, Wang et al. (2018) used gluconolactone to induce surimi myofibrillar protein to form a gel system for 3D.

A complex coacervate hydrogel is formed by the mixing of polyanions and polycations (such as alginate and polylysine), or mixing polyanions (such as chondroitin sulfate) or polycations with amphoteric polymers (Kirchmayer et al. 2015) (Fig.4). A typical example of the formation of a complex coacervate formation is the interaction between xanthan gum and gelatin to form a gel system. Gelatin is an amphoteric polymer and xanthan gum is a cationic polymer, and both of which can form a three-dimension network of gel systems through hydrogen bonding and ionic interactions (Hou et al. 2014). Cohen et al. (2009) used the interaction between gelatin and xanthan gum to form gels and incorporated various food flavors (such as strawberries, bananas, tomatoes, chocolates, etc.) for 3D printing. The print products had similar flavor and mouthfeel of many common liquid and solid foods.

In order to solve the problem of low mechanical strength and poor stability of single crosslinked gel system, some scholars used double crosslinking (two different crosslinking methods are used simultaneously in the gel preparation process) to adjust the properties (Tan et al. 2011). To create 3D printed parts used in biomedicine, a dual-cross-linking hyaluronic acid system with enough mechanical strength to be self-supportable was used for bioprinting (Ouyang et al. 2016).

Dough

For successfully 3D printing of the dough, it is generally required that the dough can be extruded smoothly and has the ability to maintain the structure and shape after the 3D printing (Yang, Zhang, Prakash, et al. 2018). However, sometimes these two conditions are contradictory: the dough with low mechanical strength can be smoothly extruded, but the shape stability is not good and easy to collapse; the dough with high mechanical strength may not be extruded. Therefore, the key to the success of 3D printing is to make the dough have appropriate physicochemical properties. As the cookie dough needs post-treatment (baking) after 3D printing, it prepared by traditional recipe might quickly deform during baking, which will affect the product quality. To solve this problem, Kim et al. (2019) added hydrophilic colloid (xanthan gum and methyl cellulose) to the cookie dough. The results showed that the dough added with 0.5%

xanthan gum had good printing precision and could maintain good structural stability during the post-treatment. With the increase of methyl cellulose, the cookie dough could be extruded smoothly with very good maintenance of shape. However, as with the control dough, regions of densely packed wheat starch particles were also partially covered with butter in the MC-added dough, so that the starch was not completely hydrated. During the post-treatment, the melting of the butter caused the dough to form loose and incomplete structure. Yang, Zhang, Prakash, et al. (2018) optimized the formulation of a baking dough and found that when the ratio of powdered sugar, butter, low-gluten flour, egg and water was 6.6:6.0:48:10.4:29, the dough had good gel and physical properties, and could be accurately printed with good shape stability. Ding et al. (2017) found that compared with the ungelatinized dough, the dough gelatinized at high temperature had higher 3D printing precision and better shape stability after being printed. Another study found that doughs containing potato starch, pea protein (1% of total), melted butter (butter: potato starch = 1:5) and water (water: potato starch = 1:1) were gelatinized at 67 °C, which had the best 3D printing accuracy and shape stability. (Feng et al. 2018). Zhang et al. (2018) used fused deposition manufacturing to study the printing properties of dough containing probiotics and found that dough prepared by mixing 30 g of water, 39 g of flour (protein content of 7.2%) and 1.17 g of calcium caseinate could stand at room temperature for 10 minutes, which was suitable for 3D printing. After the printed product was baked at 145 °C for 16 min, the survival rate of the probiotics in the products reached 10^6 CFU/g, which demonstrated that this 3D printed product could be used as a carrier of probiotics. Zhang et al. (2018) prepared a high-fiber dough prepared by mixing concentrated asparagus juice, functional carbohydrates (trehalose, high maltose) and aged flour for 3D printing. The results showed that under suitable formula and process conditions, the precision of the printed product could reach 95% or higher, and structural collapse was not occurred within 30 minutes, suggesting very good product stability. Severini, Azzollini et al. (2018) used the crushed larvae of yellow mealworms and flour to make dough. The protein and essential amino acids of the larvae powder were higher than that of the flour, so the larva powder was used to partially replace the flour to make the dough with a high nutritional value and inhibit the size reduction during the baking of the 3D printed products. This research shows that 3D printing technology can be used to manufacture innovative foods and broaden food materials such as insects.

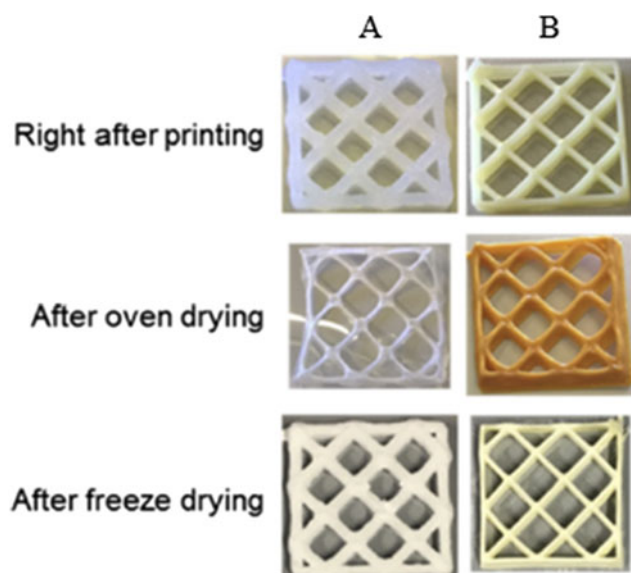


Figure 5. 3D printed representative sample before and after freeze-drying (Lille et al. 2018). (A) 1.5% Cellulose Nanofiber + 5% starch (initial solid content $\leq 35\%$); (B) 0.8% Cellulose Nanofiber + 50% semi-skimmed milk powder (initial solid content $\geq 45\%$).

Post-treatment

After the completion of 3D printing, there may have some defects on the surface of the product, which requires post-processing operations to improve printing precision and shape stability. For 3D printed non-food products, the post-processing techniques are relatively mature, which include support removal, model repair, polishing, surface painting, and coloring etc. However, the post-processing of 3D printed food products is completely different, which mainly includes drying, cooking, cooling, and 4D printing.

Drying

The drying methods currently used for post-processing of 3D printed products are freeze drying, oven drying and vacuum microwave drying. Lille et al. (2018) compared the effects of oven drying (at 100°C for 20–30 min) and freeze drying (-18°C) on the shape stability of 3D printed products that are rich in protein and cellulose. The results showed that the printed samples with lower initial dry matter (initial dry matter $\leq 35\%$) can keep the structure more stable by freeze drying; for samples with high initial dry matter (initial dry matter $\geq 45\%$), there was no shape difference after the two drying methods, partly because of higher initial structural strength when the water content was lower (Fig. 5). Yang et al. (2018) mixed potato starch and concentrated mango juice at a ratio of 13.04:86.96 to make a juice gel for 3D printing. The printed product was dried by vacuum microwave drying at 150 W for 4 min, and the shape stability was very well maintained, in which the precision of the sample shape was up to 99.8%. In addition, the sensory quality of the product was acceptable. The research suggested that vacuum microwave drying could be a promising method for maintaining the shape stability (Fig. 6) and sensory quality of 3D printed food products. This method has

the potential to be applied for post-treatment of other 3D printed gel materials.

Rapid cooling

Rapid cooling is another method to effectively maintain the structure stability of 3D printed products. For example, Lipton et al. (2010) observed that the shape of printed pastry was effectively retained after fast frozen. The inner interlayer of the pastry was also effectively retained even after the subsequently baking operation. Yang et al. (2018) quickly cooled the 3D printed dough samples in an ultra-low temperature freezer (-65°C) for 0, 5, 10 min (Fig. 7). The result showed that without the frozen process, the baked dough was collapsed before baking. However, after fast frozen for 5–10 minutes, the baked dough had a very good shape and structure stability, especially the sample with fast frozen for 10 minutes, which matched the structure of the initial design.

Cooking

Traditional cooking

To promote the commercialization and consumer acceptance of 3D printed foods, 3D printing should be adaptable to traditional food processing techniques, for instance, baking, steaming, frying and other cooking methods. However, to maintain the shape stability during cooking is a big challenge to 3D printed foods (Lipton et al. 2015). There are two main ways to solve the shape stability of printed products during cooking: formulation control and additives. Generally, the formulation of the 3D printing food is critical to the shape stability during cooking. Severini, Derossi, and Azzollini (2016) mixed 100 g wheat flour with 54 g distilled water to make the dough, and then allowed to stand for 30 min before 3D printing. After baked at 200°C for 15 min, the product shape accuracy was very close to the target structure (Fig. 8). However, due to physical and chemical changes such as protein denaturation, dehydration, and starch gelatinization during baking, the sample surface after baking was rough. In addition, studies have shown that the addition of transglutaminase ($\geq 0.5\%$) to the meat slurry allows the printed sample to remain shape stability (Lipton et al. 2010; Lipton et al. 2015). For example, transglutaminase was added to the scallop meat paste for 3D printing, and the fried product retained most of the original shape, with very thin areas of deformation (Fig. 9) (Lipton et al. 2010; Lipton et al. 2015).

Modern cooking techniques

Modern cooking techniques use advanced technologies such as information technology and mechatronics to achieve new taste and flavor (Fukuchi et al. 2012). Unlike traditional cooking methods (e.g. baking, cooking, frying), in which all raw materials are uniformly heated, modern cooking technique of laser cooking can locally heat part of the raw materials in a short time, resulting in a new taste of food and

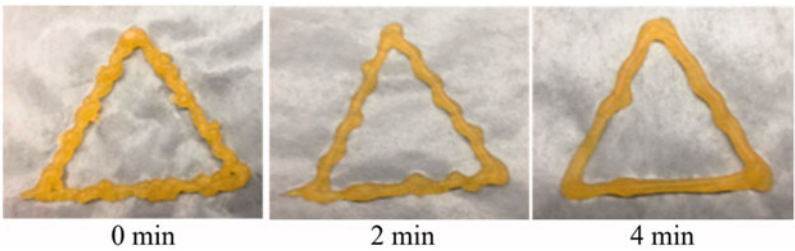


Figure 6. Representative photos of 3 D printed mango juice gel samples with MVD post-treatment 20 min after printing (Yang, Zhang, and Liu 2018).

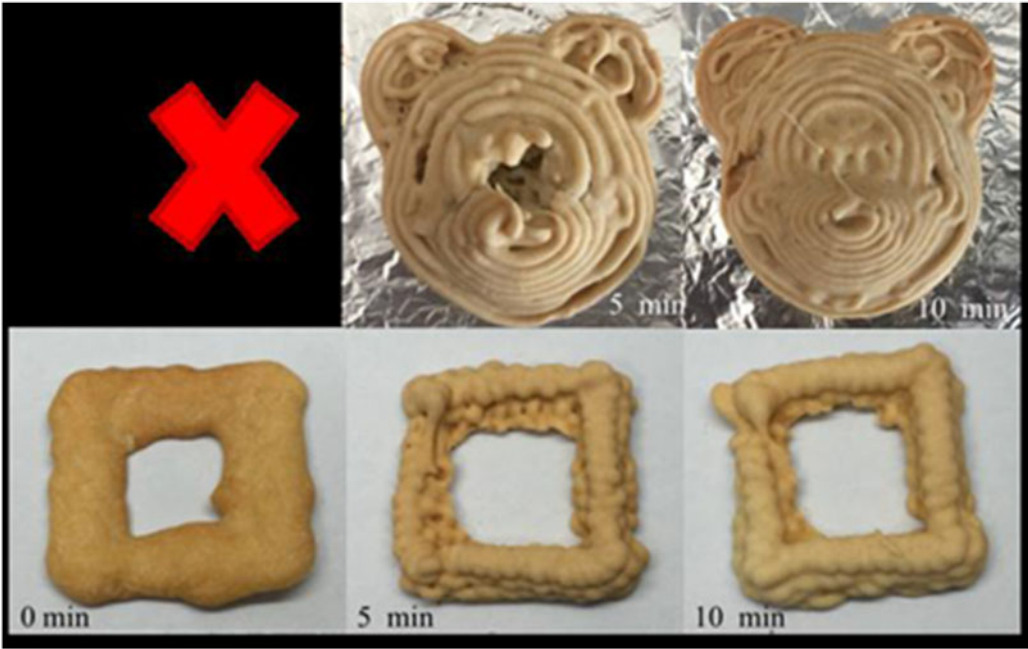


Figure 7. Effect of different fast cooling times on printed baking dough before baking (Yang, Zhang, Prakash, et al. 2018).

Designed objects	Printed raw objects	Printed cooked objects

Figure 8. Shape of 3 D printed cereal-based samples before and after baking (Severini, Derossi and Azzollini 2016).

eating experience (Fukuchi et al. 2012). A Japanese researcher connected a laser cutter to a computer equipped with image processing software and a camera, took pictures of the raw bacon with a camera, and then used image analysis software to create a heat pattern from the camera image by detecting the fat portion of the bacon. Finally, the heat pattern was sent to the laser cutter, the cutter could cook the fat in the bacon according to the heat pattern, thereby obtaining the “melt-fat raw bacon”, so that people could enjoy the two different flavors of cooked fat and raw bacon (Fukuchi et al. 2012). In recent years, researchers have combined 3 D printing and laser cooking to process food. For instance, a researcher at Columbia University used two servo-driven mirrors to reflect the laser light onto the

printed food, which could be cooked in intricate patterns of precise heat (Cameron 2018). Blutinger et al. (2018) printed the dough into cuboids with a thickness of 3.0 mm and length and width of 30 mm respectively. The printed dough was processed using two different cooking methods (baking and laser cooking). The results showed that the laser cooked and oven baked dough samples had very similar starch swelling and nutritional levels. By changing the laser cooking mode, one could control texture of the final cooked product, making the product more acceptable to consumers. Therefore, laser cooking is also a very promising post-processing method for 3 D printed food product.

In addition, vacuum cooking (or “sous vide”) is another modern cooking method. The food materials are

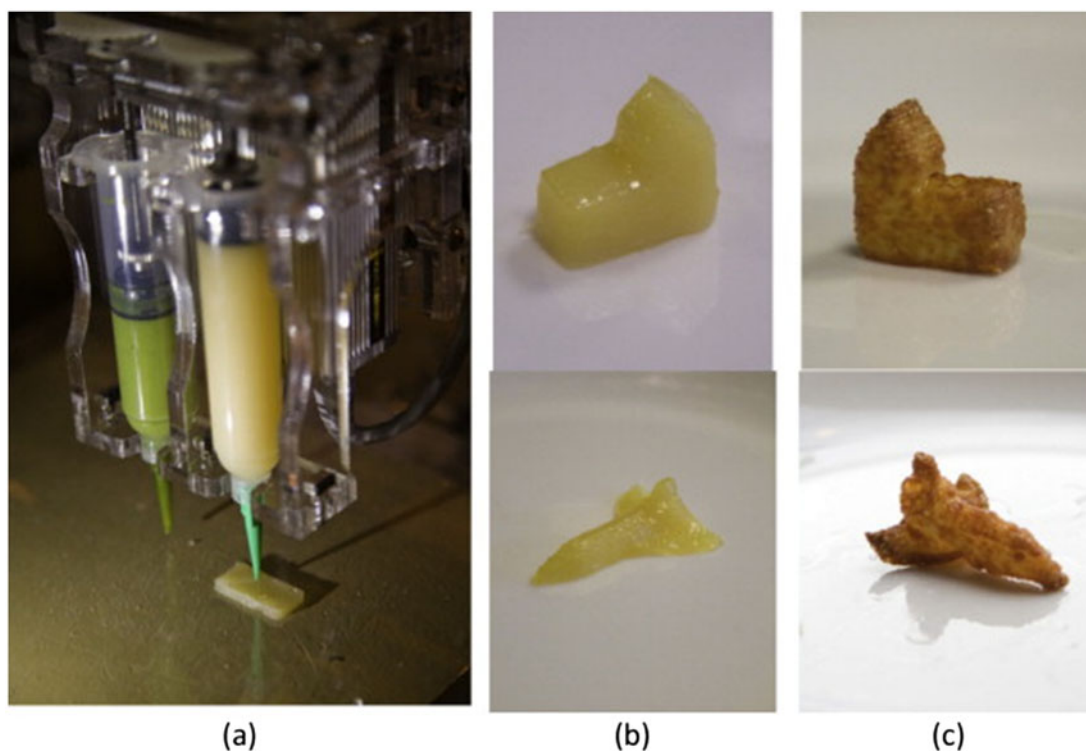


Figure 9. (A) 3D printing of scallop meat; (B) and (C) are 3D printed samples before and after frying, respectively (Lipton et al. 2015).

vacuum-packed in a heat-stable vacuum bag and heated for a long time at a low temperature (Fukuchi et al. 2012; Lipton et al. 2015). Compared with traditional cooking methods, Sous vide can inhibit the oxidation of some substances to avoid peculiar smell, prevent the volatilization of flavor and moisture during cooking, and improve the shelf life of food (Baldwin 2012). Lipton et al. (2010) added transglutaminase to turkey purees for 3D printing. The printed product was vacuum-cooked and the overall shape was preserved, although the meat was a bit shrunk inward and bent. This study indicated that post-processing of 3D printed products is very important to maintain the shape stability of the food.

4D Printing

4D printing is developed on the basis of 3D printing. It is used to describe the combination of 3D printing technology with smart materials, i.e. using smart materials as the object of 3D printing (Shin, Kim, and Kim 2017). After being printed and exposed to predetermined stimuli (such as heat, osmotic pressure, light, etc.), the printed product can self-transform in shape and function to form a new configuration, which is termed as the fourth dimension printing, i.e. 4D printing (Miao et al. 2017; Shin, Kim, and Kim 2017). 4D printing has received extensive attention since its introduction, and has been widely used in material science, manufacturing industry, bioengineering, and medical fields (Momeni, Liu, and Ni 2017). Wang et al. (2017) first reported 4D printing of food. They printed ordinary food materials (protein, cellulose or starch) into edible 2D

films that would twist and curl in a unique way to form 3D foods when the 2D films were cooked. This study facilitated the 4D printing of food, and it is expected that the application of this technology in food processing has great potential (Fig.10).

Challenges of food 3D printing technology and prospects

In the last decade, great efforts have been made globally aiming at applying 3D printing into the food industry, mainly focusing on the utilization of new food sources (Severini, Azzollini et al. 2018; Sun, Zhou, Huang et al. 2015), development of personalized food (Sun et al. 2018), optimization of printing parameters (Liu et al. 2017) and designing of printing equipment (Le Tohic et al. 2018; Malone and Lipson 2007). However, the broad application of this technology in the food industry still presents many difficulties due to several reasons:

First, the restriction of food materials. There are very few food materials that can be printed directly, such as cheese (Kern, Weiss, and Hinrichs 2018; Le Tohic et al. 2018), dough (Yang, Zhang, Prakash, et al. 2018), hydrogel (Liu, Bhandari, Prakash, Mantihal et al. 2019), chocolate (Mantihal et al. 2017). Most of the food ingredients are not directly printable, such as meat, fruits and vegetables (Sun, Zhou, Huang et al. 2015). To expand food 3D printing, it is necessary to preprocess the food materials to have suitable physical and chemical properties to be printable. Food materials used in 3D printing have limited shelf life and their rheological properties may change during storage, which will



Figure 10. 4 D printed deformed dough (Wang et al. 2017).

in turn affect the printability. Moreover, the materials that are currently used for 3 D printing are generally soft materials such as powders, pastes, gels, and doughs, etc. When ordinary foods are changed into these states for 3 D printing, inevitably, food loses some nutrition value and sensory qualities. Research in the pretreatment technology that makes food material printable but with minimal nutrient loss, could be a new research area for food 3 D printing.

Second, the pretreatment technology of food materials for 3 D printing is also limited. In the current research, only some traditional methods such as comminution and gelation are used for this purpose, which are generally simple and inefficient. The application of new and efficient technologies such as microwave treatment and ultrasonic treatment before 3 D printing needs further study.

Third, challenge of food 3 D printing post-processing technology. Only a small portion of 3 D printed food does not require a post-treatment process, such as 3 D printed chocolate. Most of other 3 D printed food products require post-processing such as baking, frying, cooking, drying, and cooling. However, the post-treatment process sometimes affects the sensory properties of food. It is advantageous. For example, after baking, the pizza that was made by Bee Hex 3 D printer tasted like a normal pizza at the beginning and then closer to crackers after a few minutes because of its ultra-thin crust, which gave people a new taste experience (Sun et al. 2018). But sometimes it is unfavorable, such as the loss of nutrients, the formation of undesirable flavors, changes in the color of the product, and so on (Dankar et al. 2018). In addition, there are very few studies on post-processing techniques for food 3 D printing. Therefore, the development of new post-processing techniques, as well as avoiding the adverse effects of post-processing, are issues that need to be studied in the future.

Fourth, lack of research in 3 D printing of functional foods. The current research is still mainly focused on the research of printable materials and optimization of processing parameters. There are few researches in 3 D-printing of personalized and functional foods for specific cohort of people, which is an important factor limiting the development of food 3 D printing.

In addition, potential food safety issues and consumer acceptance of 3 D printed foods are also important but the research in these topics is scarce (Dankar et al. 2018).

Conclusion

In summary, the pretreatment of food materials and the post-treatment of printed products are the main factors affecting the accuracy of food 3 D printing and the shape stability of printed products. For food 3 D printing, only very few food materials can be directly used for printing such as cocoa powder, powdered sugar, etc. Most materials require proper pretreatment (such as comminution, gelation, preparation of dough, etc.) according to the states and characteristics of the materials, so that the materials used for printing have the proper rheology so as to be easily extruded from the nozzle, as well as appropriate mechanical properties to maintain the shape fidelity of the printed product. Compared with the pretreatment of materials, the post-processing of food 3 D printing should not be underestimated. In order to maintain the stability of the printed product structure, it is often necessary to dry or freeze the printed product. However, since the object of 3 D printing is food, in order to make the 3 D printed food acceptable to the public, it is often necessary to cook like a normal food to make it acceptable to the public. However, the pretreatment and post-treatment methods involved in food 3 D printing are simple and inefficient, and the development of new and efficient methods is one of the challenges of rapid development of 3 D printing in the food industry.

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